Satellite Retrievals → Global Assimilation → Regional Prediction → Validation

RAQMS Regional Air Quality Modeling System

INTEX-NA

RAQMS chemical and aerosol studies during the 2004 NASA INTEX-NA field campaign

R. Bradley Pierce[1], Jassim Al-Saadi[1], Chieko Kittaka[1], Duncan Fairlie[1], Todd K. Schaack[2], Gretchen Lingenfilser[1], Donald R. Johnson[2], Tom H. Zapotocny[2],Allen J. Lenzen[2], Matt Hitchman[3],Greg Tripoli[3],Marcus Buker[3]

<u>Insitu data provided by:</u>

M. Avery (LaRC), Anne Thompson, (Penn State), R. Cohen (UC Berkley), J. Dibb (UNH)

<u>Boxmodel results provided by:</u>

J. Crawford (LaRC)

<u>Satellite data provided by:</u>

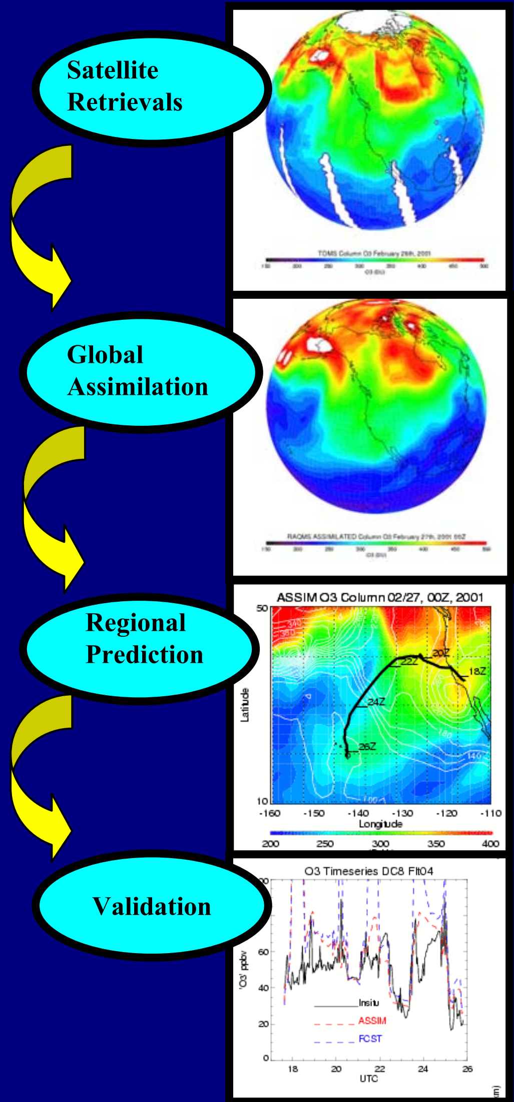
R. McPeters (GSFC), Allen Chu (GSFC), Didier Rault (LaRC), R. Martin (Dalhousie)

<u>EPA Surface data provided by:</u>

J. Szykman (USEPA)

[1] NASA Langley Research Center
[2] University of Wisconsin, Space Science and Engineering Center,
[3] University of Wisconsin, Atmospheric and Oceanic Sciences

Regional Air Quality Modeling System (RAQMS) Ozone Assimilation/Prediction February 27, 2001

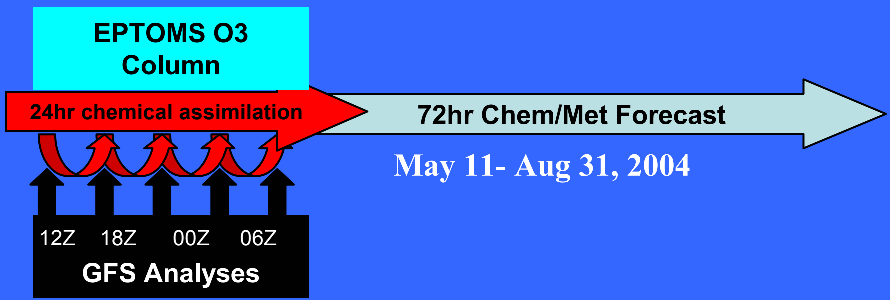


RAQMS [Pierce et al., 2003] is a nested global-to regional-scale meteorological and chemical modeling system for assimilating and predicting the chemical state of the atmosphere (air quality).

RAQMS$_G$ Chemical Assimilation Chem/Met Forecast Cycle

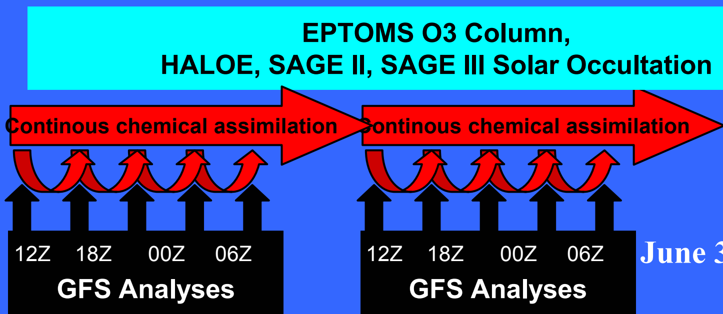
Real-Time

2x2.5 Degree resolution
Kawa Table photolysis
Climatological wet deposition



Post-mission

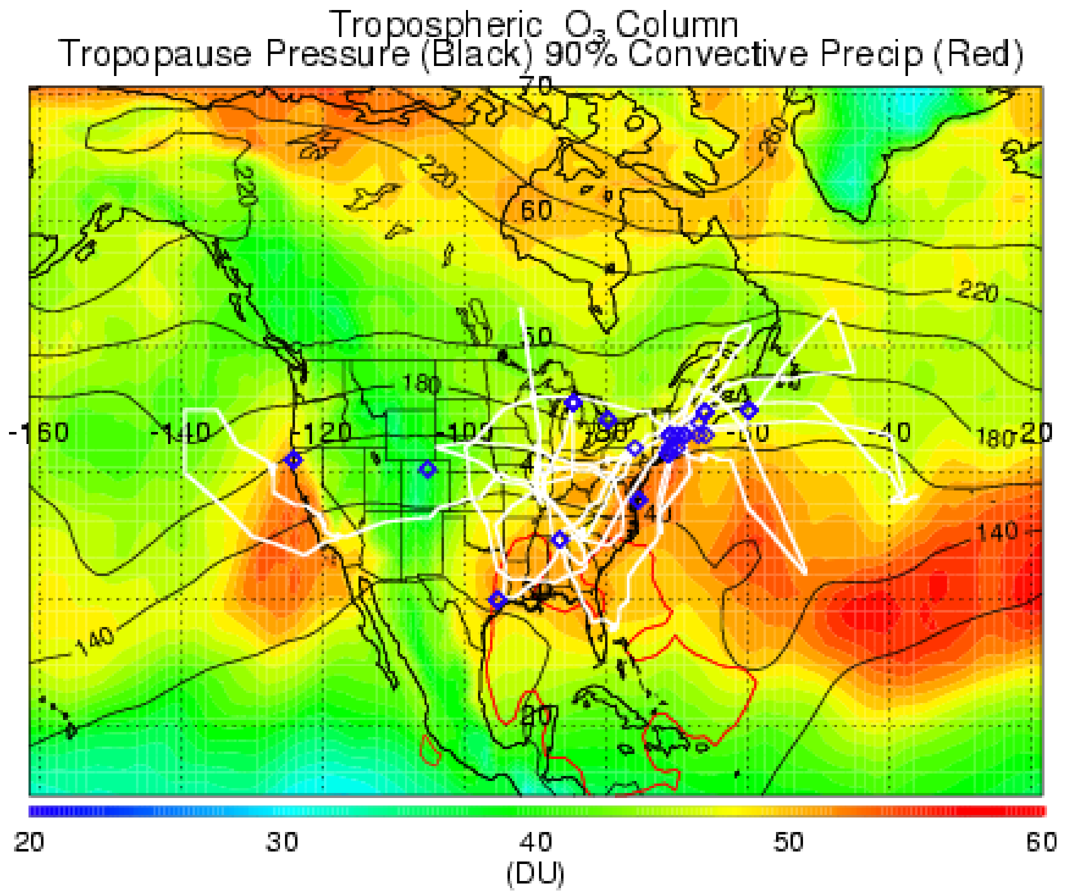
1.2x1.4 Degree resolution
Fast-J2 photolysis
GMI (Harvard) wet deposition



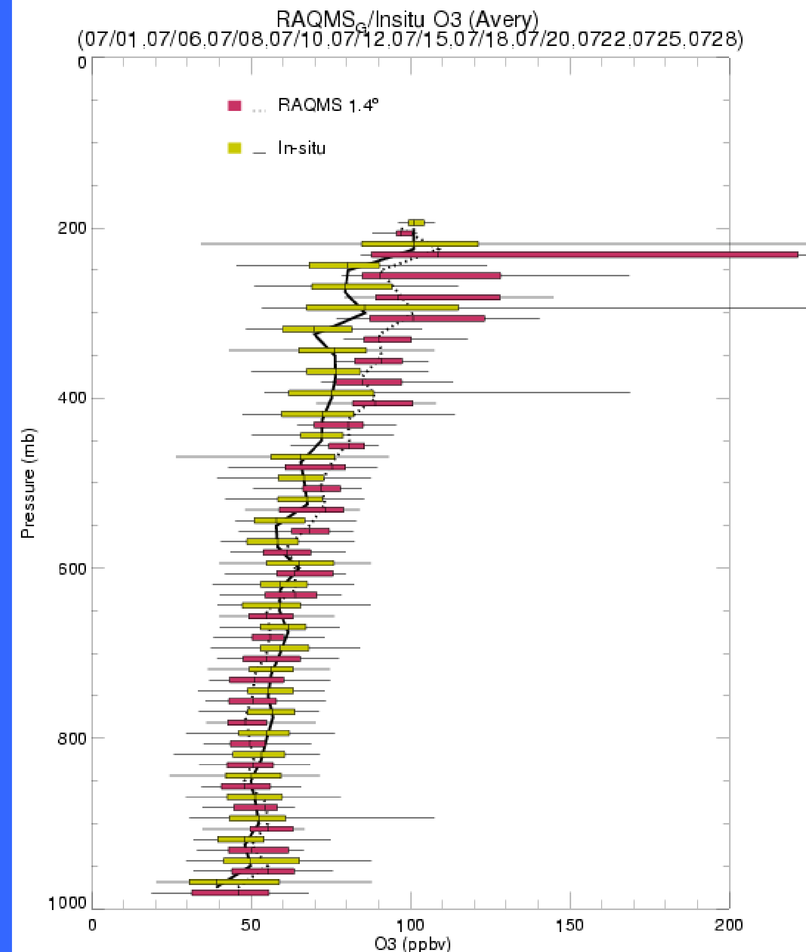
June 30-July 31, 2004

Global component of the LaRC/UW-Madison Regional Air Quality Modeling System (RAQMS) uses the UW-Hybrid dynamical core, LaRC unified strat/trop chemistry, and Statistical Digital Filtering (SFD) for real-time TOMS Ozone assimilation and chemical/dynamical predictions and post mission TOMS+Solar Occultation assimilation.

Verification of RAQMS Upper Air O3 Analysis: INTEX RAQMS/DC8 Insitu O3 (M. Avery, LaRC)

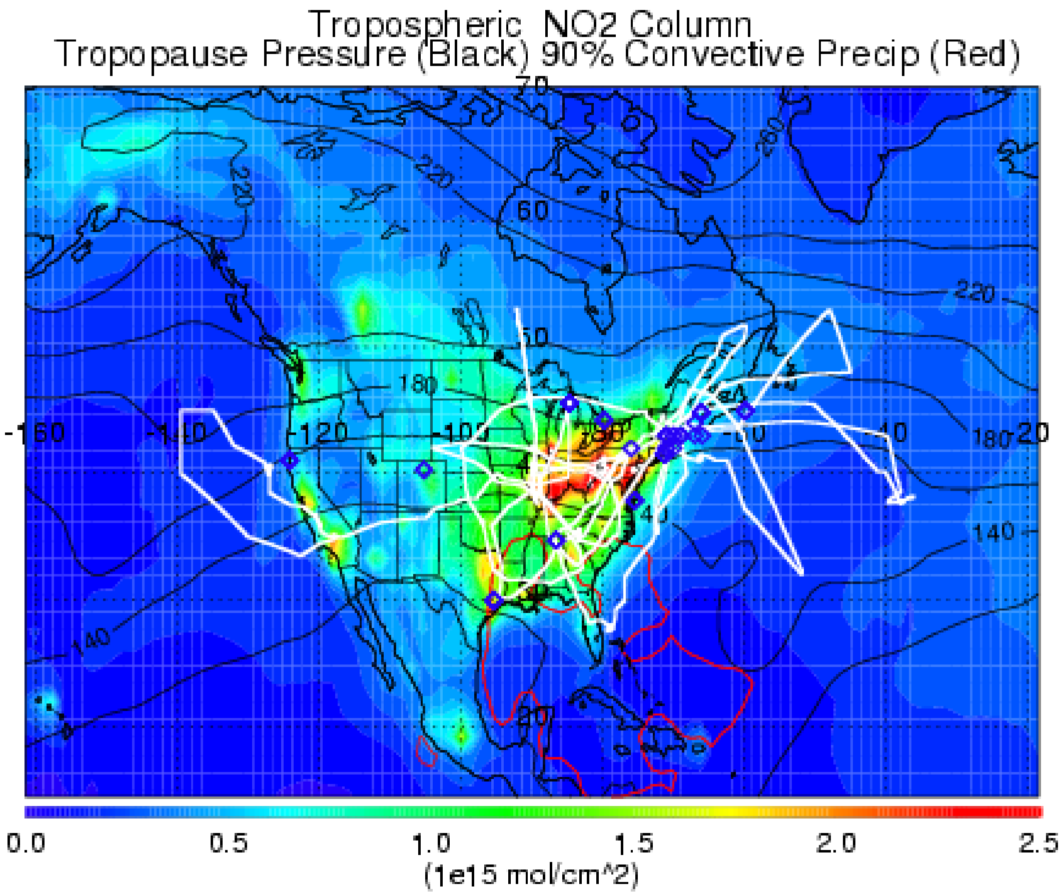


July 2004 RAQMS TOC and DC8 Flight Tracks

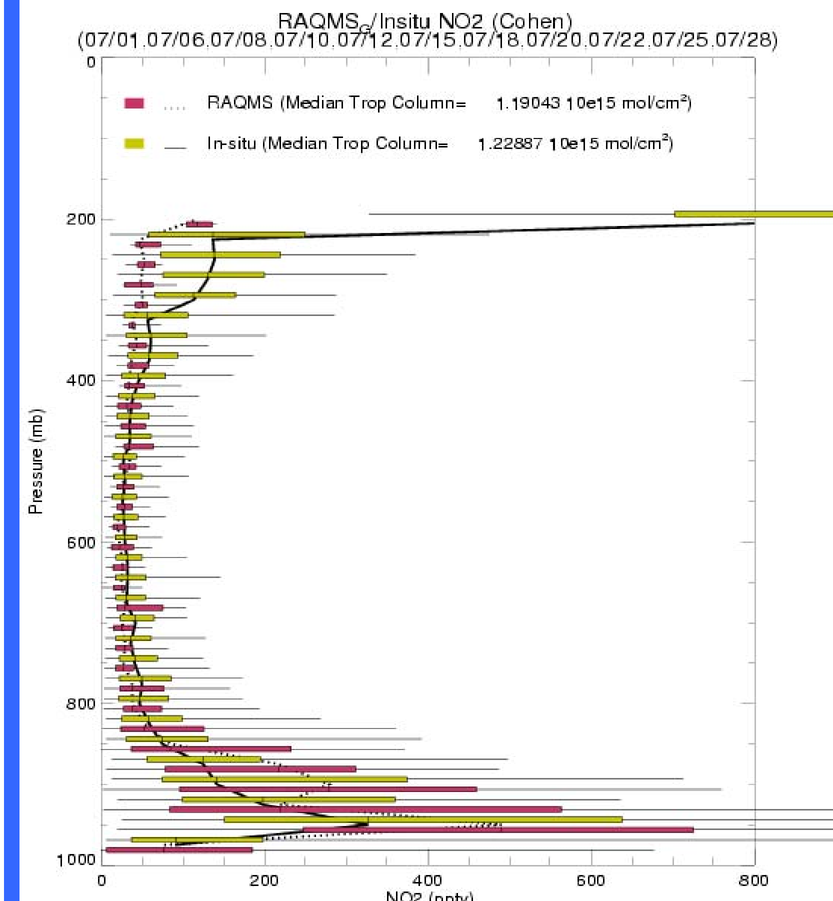


RAQMS O3 analysis shows very good agreement with in-situ O3 except for overestimates associated with tropopause encounters

Verification of RAQMS Upper Air NO2 Analysis: INTEX RAQMS/DC8 Insitu NO2 (R. Cohen, UC-Berkley)

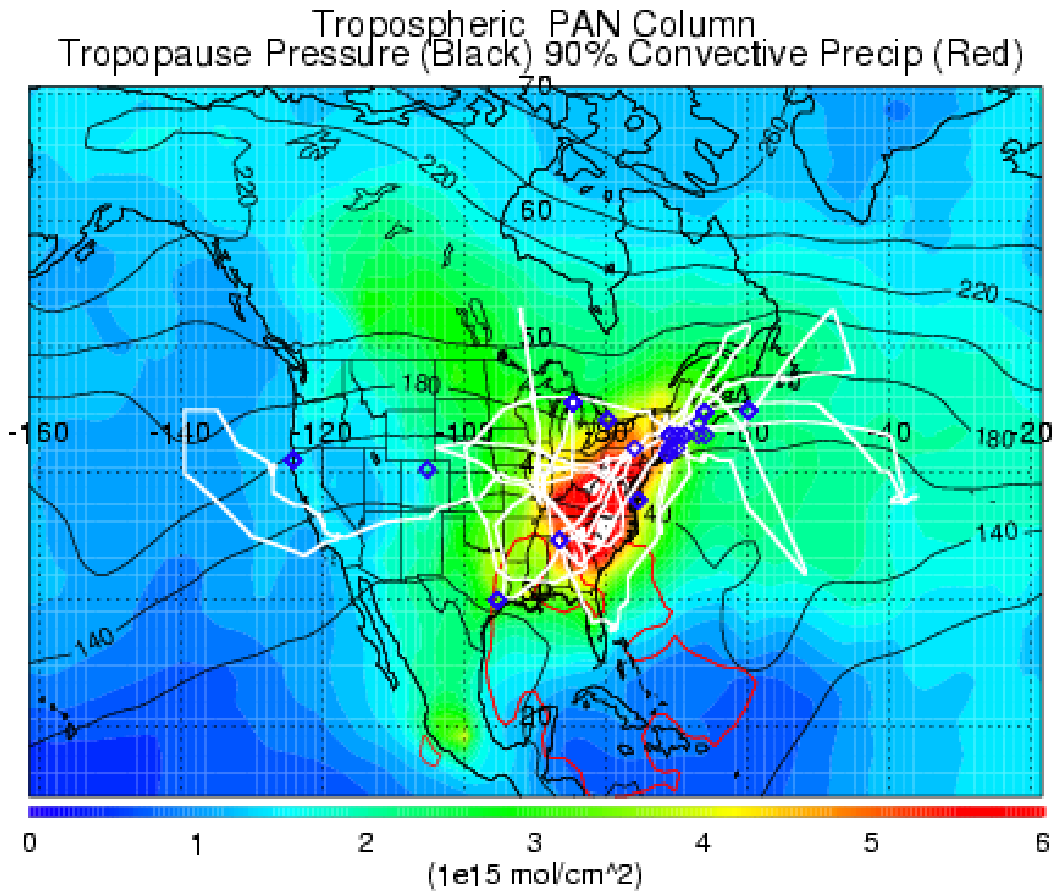


July 2004 RAQMS NO2 and DC8 Flight Tracks

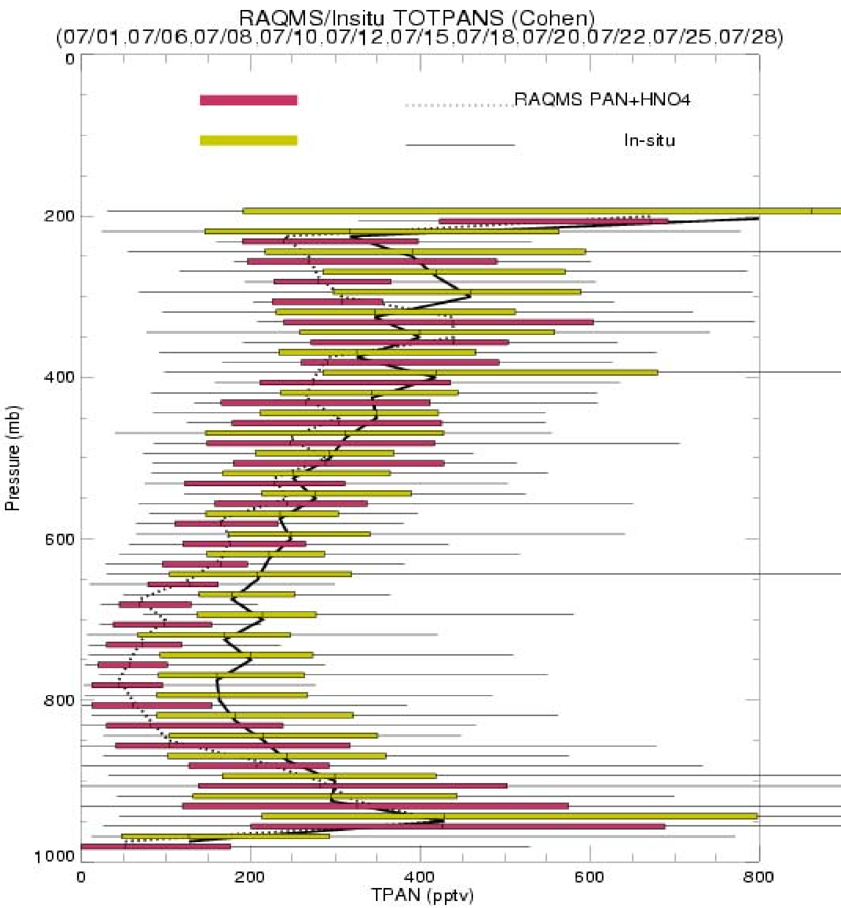


RAQMS NO2 underestimates upper tropospheric NO2 (lightning NOx ?), and overestimates PBL median mixing ratios. RAQMS NO2 column is within 5% of the observed median column.

Verification of RAQMS Upper Air PAN Analysis: INTEX RAQMS/DC8 Insitu TPAN (R. Cohen, UC Berkley)

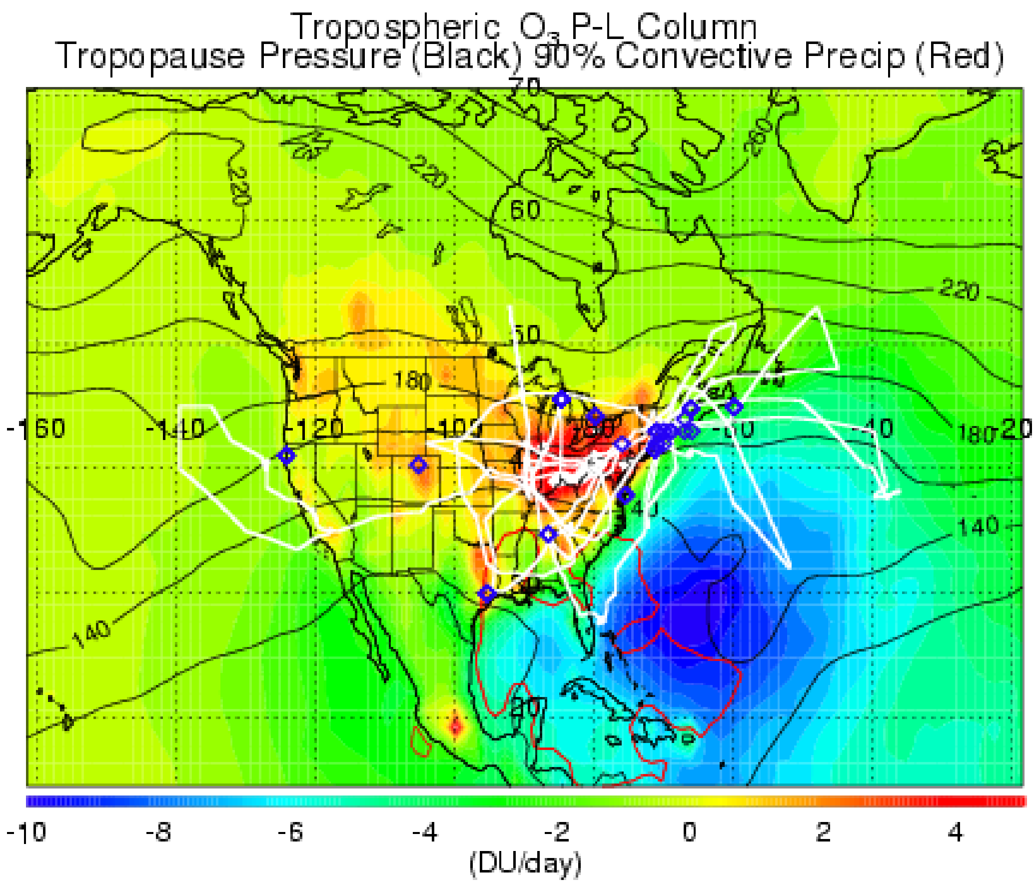


July 2004 RAQMS PAN and DC8 Flight Tracks

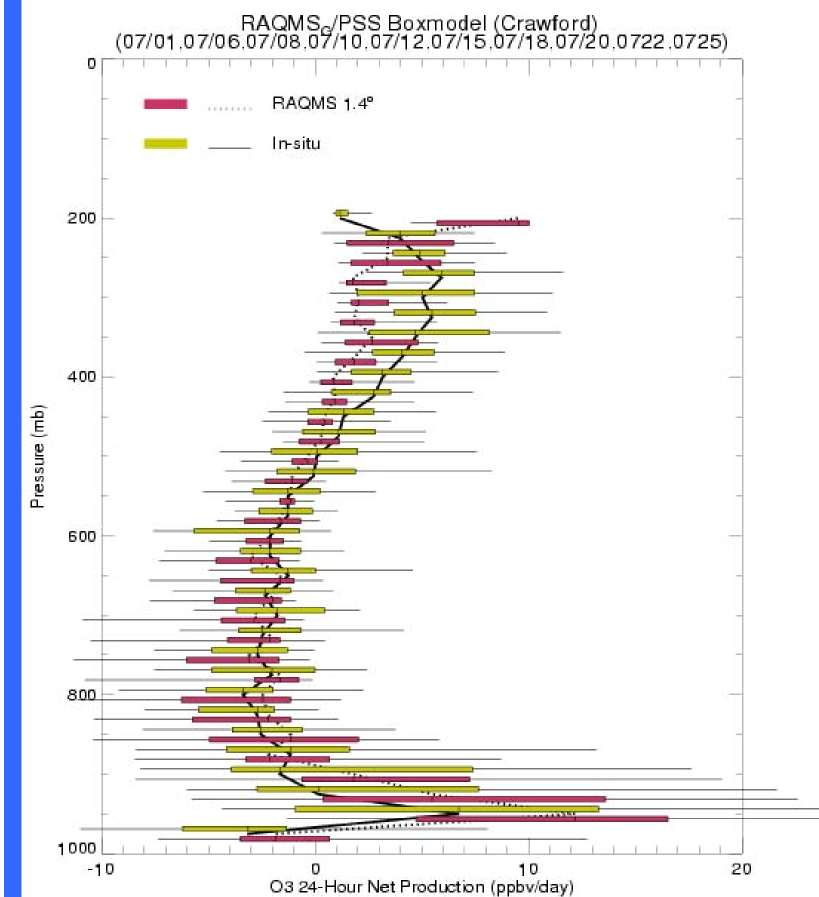


RAQMS PAN captures the observed profile shape (which indicates strong convective influences) of the observed PAN, but tends to underestimate the mixing ratio above the PBL.

Verification of RAQMS Upper Air O3 P-L Analysis: INTEX RAQMS/DC8 Boxmodel P-L (Crawford, LaRC)

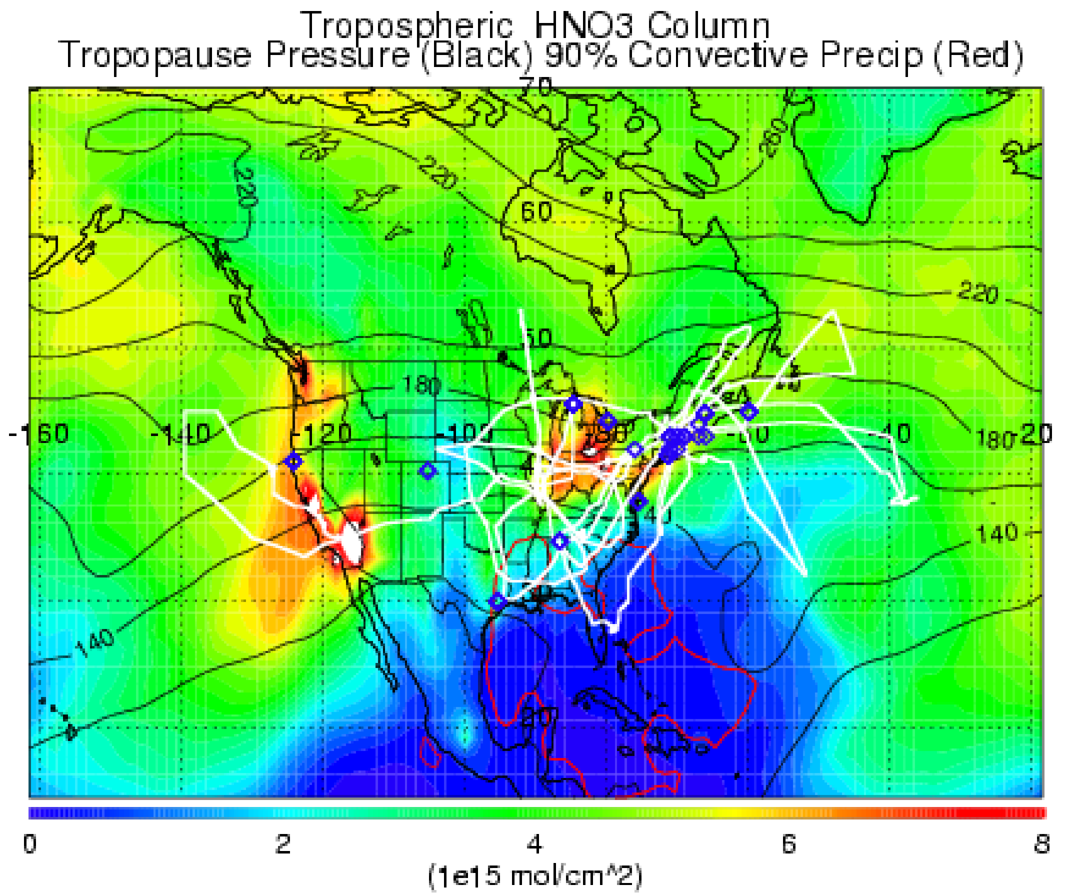


July 2004 RAQMS O3 P-L and DC8 Flight Tracks

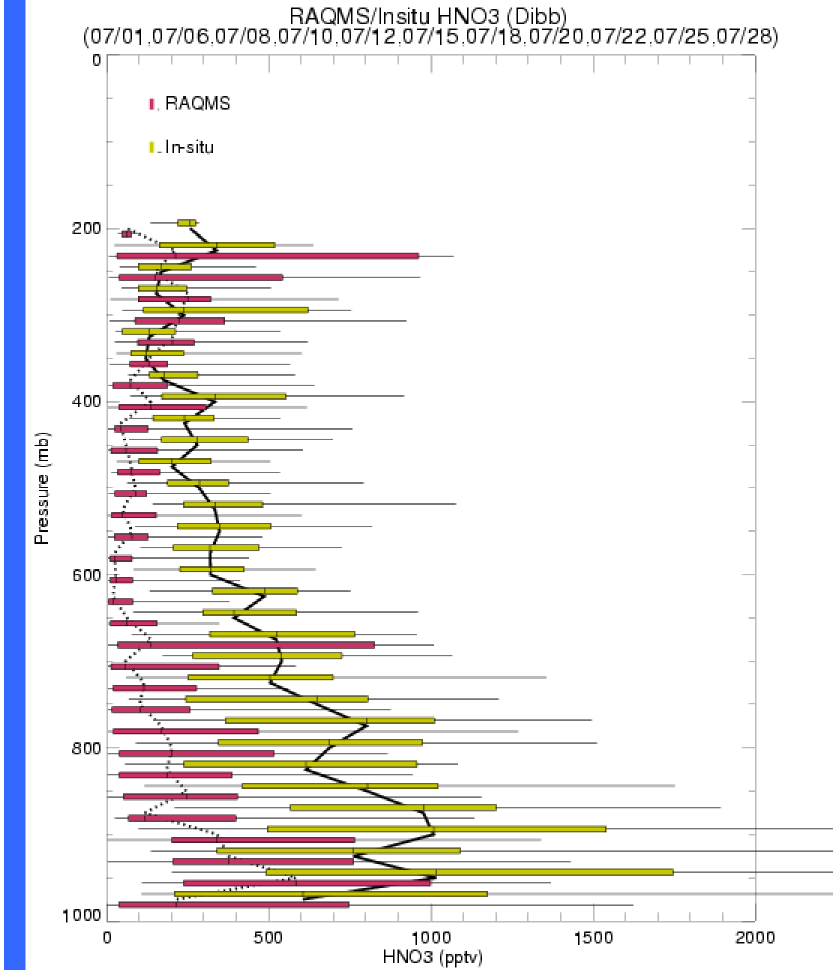


RAQMS O3 P-L analysis shows good agreement with constrained Box model calculations in the middle troposphere. RAQMS P-L underestimates upper tropospheric (lightning NOx ?), and overestimates PBL P-L.

Verification of RAQMS Upper Air HNO3 Analysis: INTEX RAQMS/DC8 Insitu HNO3 (J. Dibb, UNH)



July 2004 RAQMS HNO3 and DC8 Flight Tracks

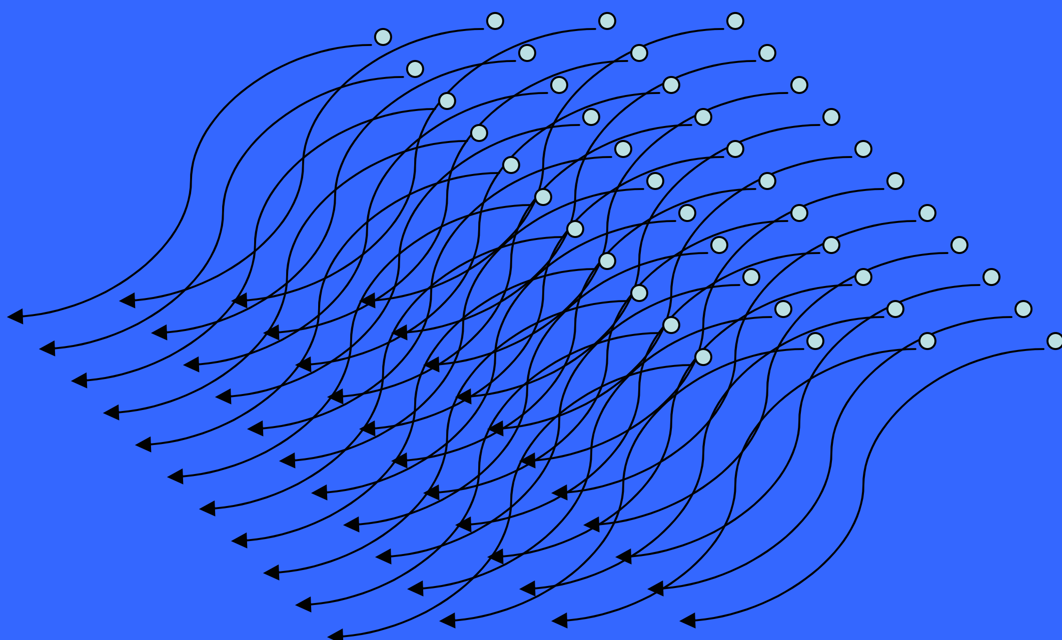


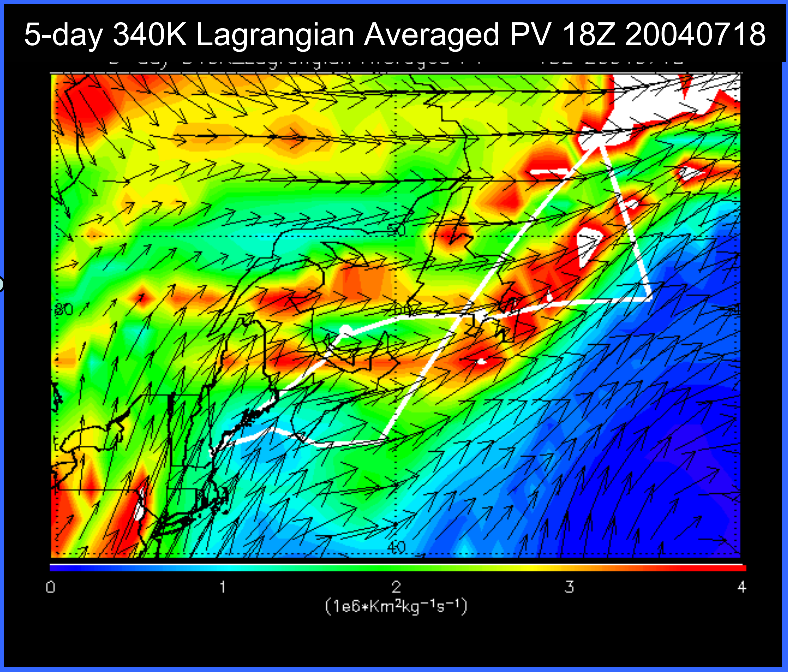
RAQMS HNO3 underestimates observed HNO3 below 400mb with GMI implementation of Harvard wet deposition.

RAQMS Lagrangian Analyses: (D. Fairlie Lead)

Method:

- Initialize uniform 3D grid of trajectories
- Compute backward trajectories
- Sample and average RAQMS chem/dyn fields along back trajectories.
- Map Lagrangian average back onto original uniform grid.



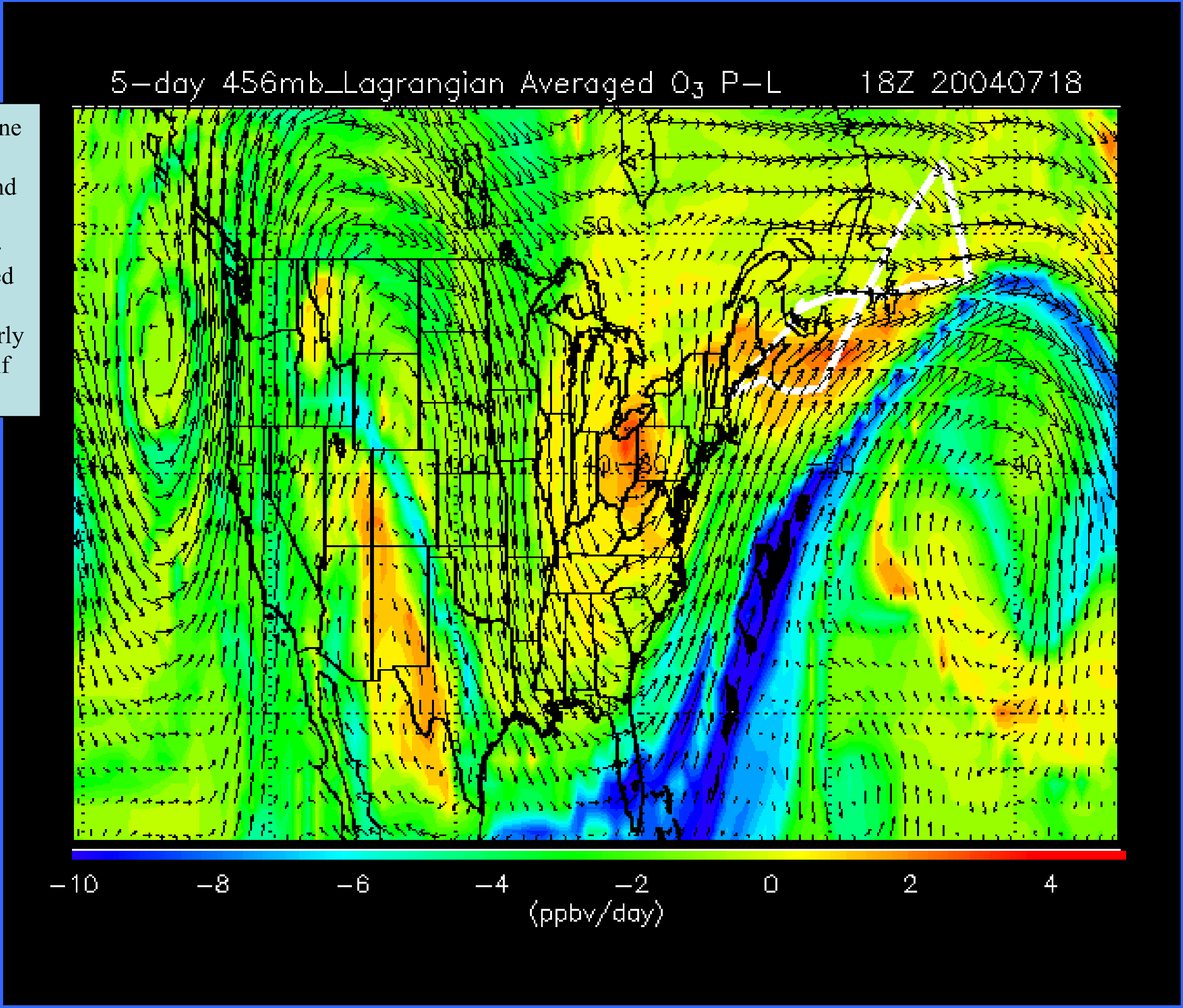


Result:

Synoptically mapped photochemical, mixing, and transport processes following air parcels via Reverse Domain Filling (RDF)

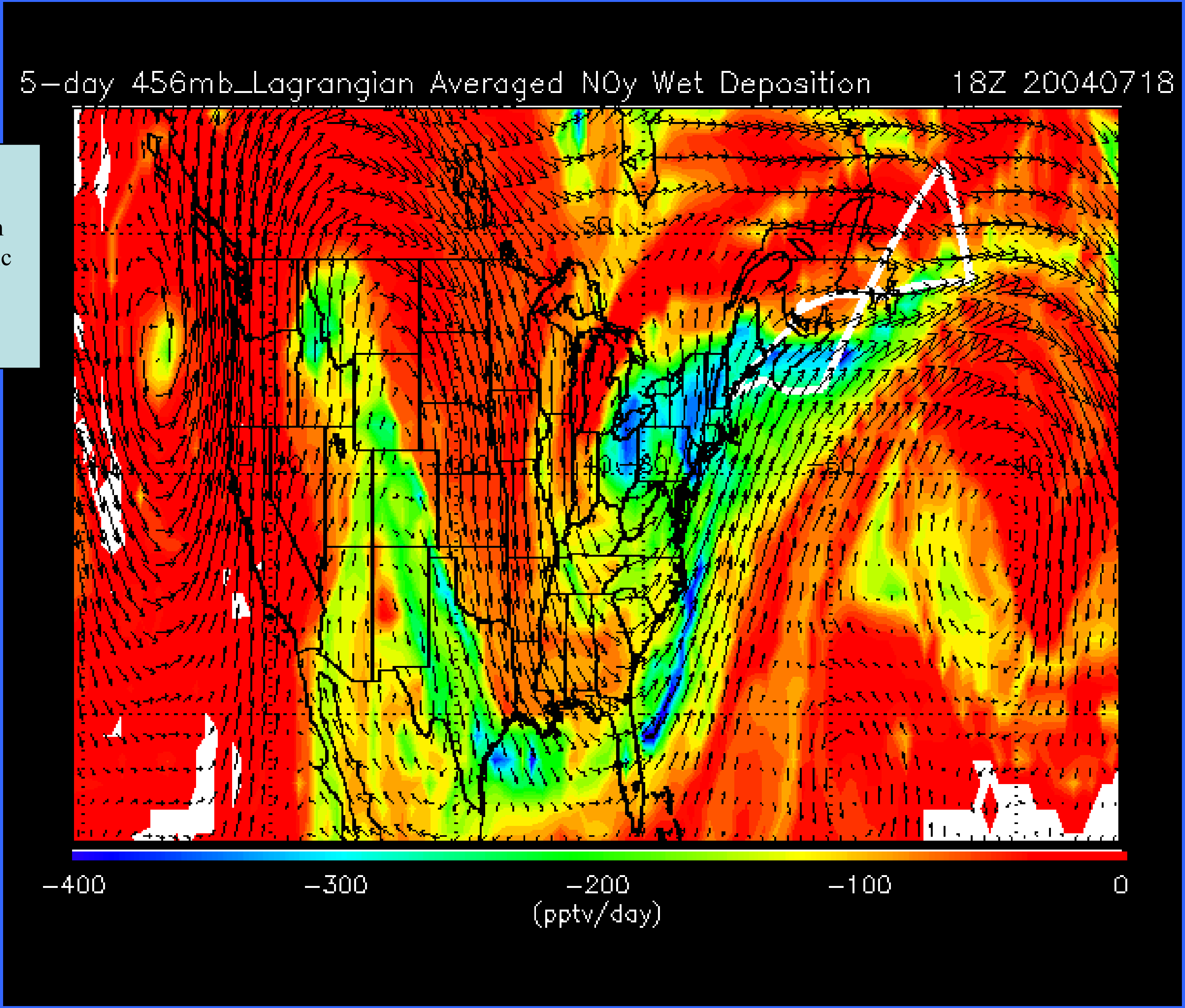
RAQMS$_G$ 456mb Lagrangian Analyses 18Z July 18th, 2004

Enhanced ozone production in NE outflow and within trough axis over mid-west. Enhanced ozone loss within Southerly flow from Gulf of Mexico



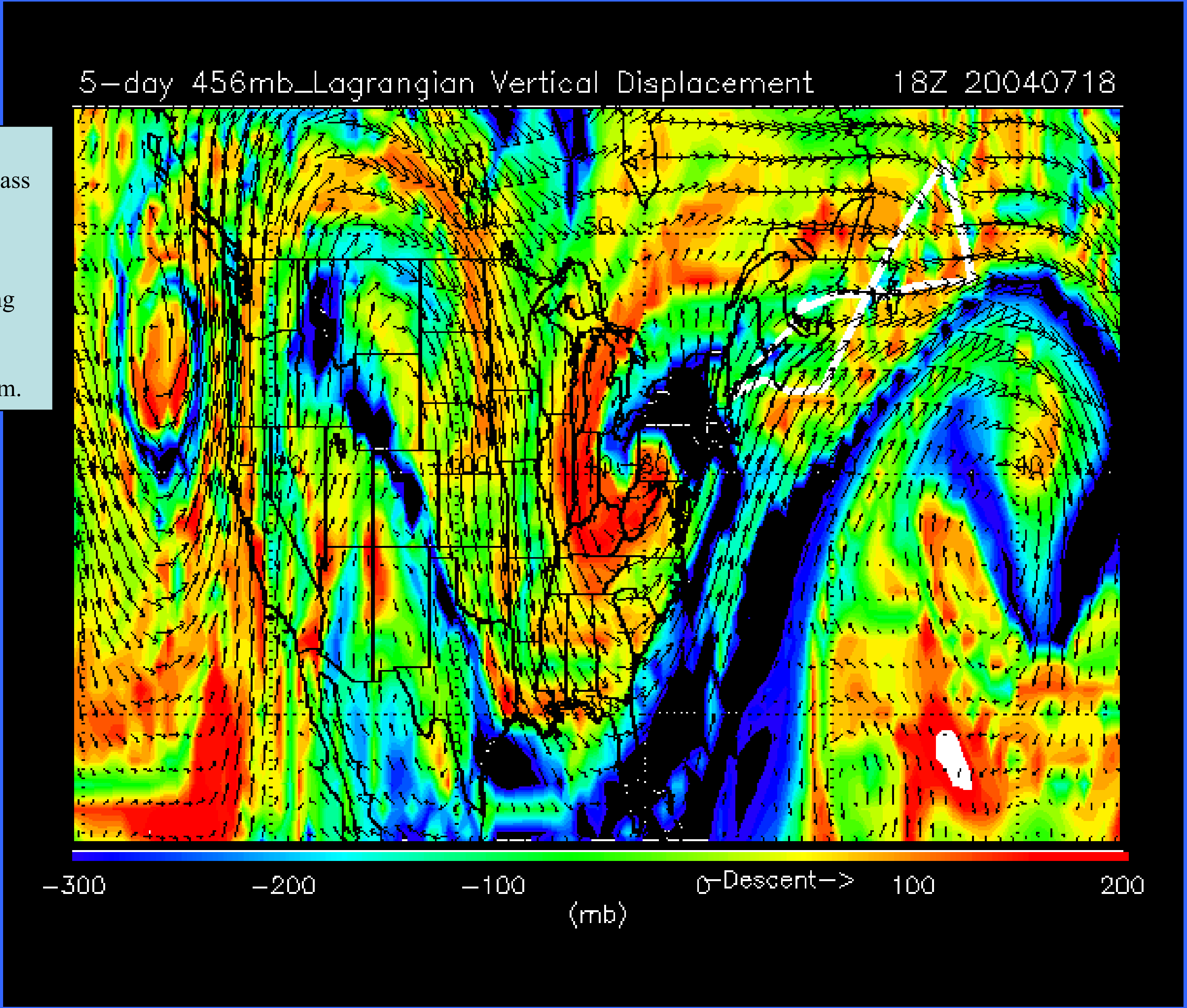
RAQMS$_G$ 456mb Lagrangian Analyses 18Z July 18th, 2004

Enhanced loss of NOy due to wet deposition within synoptic and frontal precipitation bands.

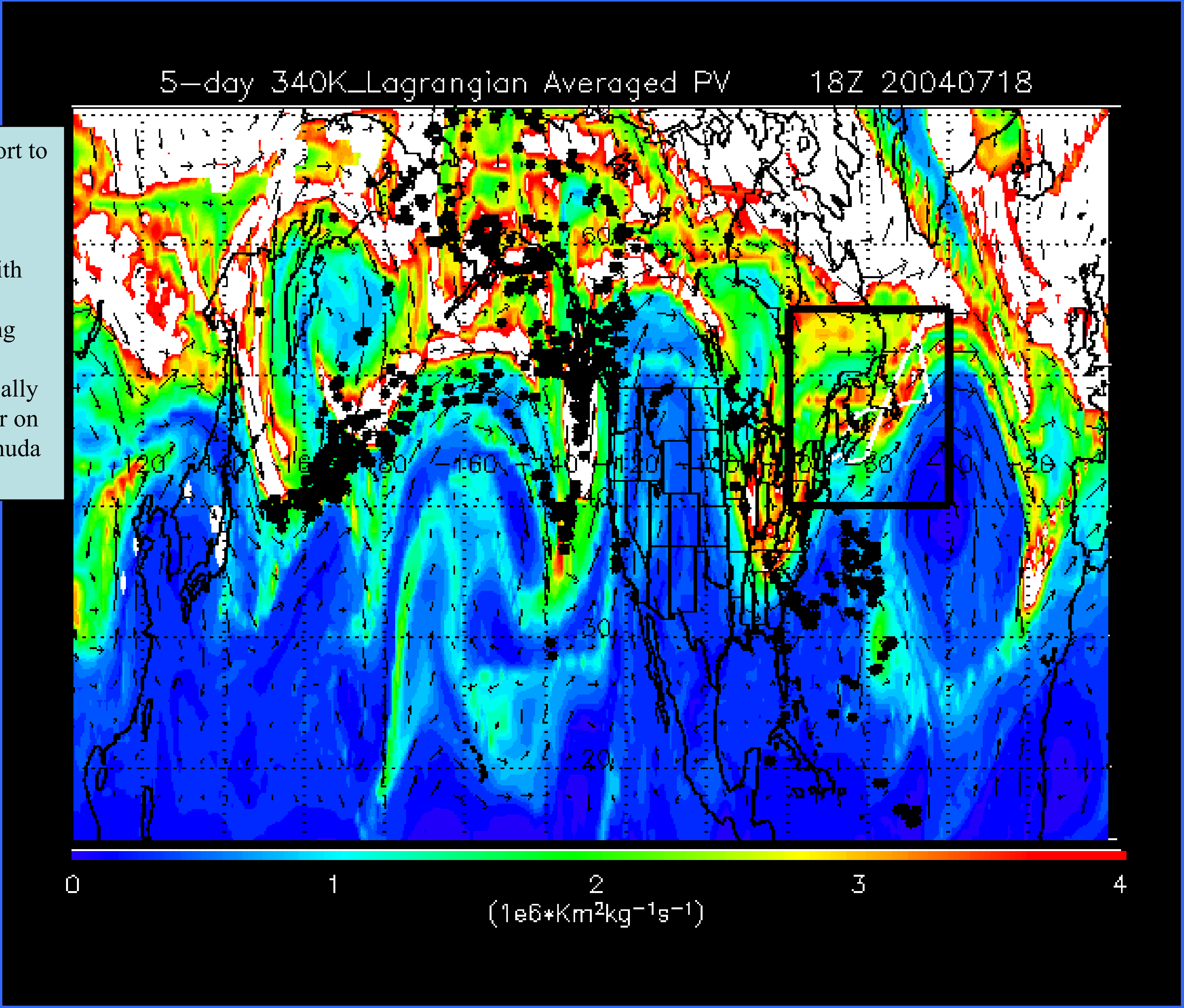


RAQMS$_G$ 456mb Lagrangian Analyses 18Z July 18th, 2004

Ascent of maritime airmass from Gulf of Mexico. Descending airmass coming in behind synoptic low pressure system.



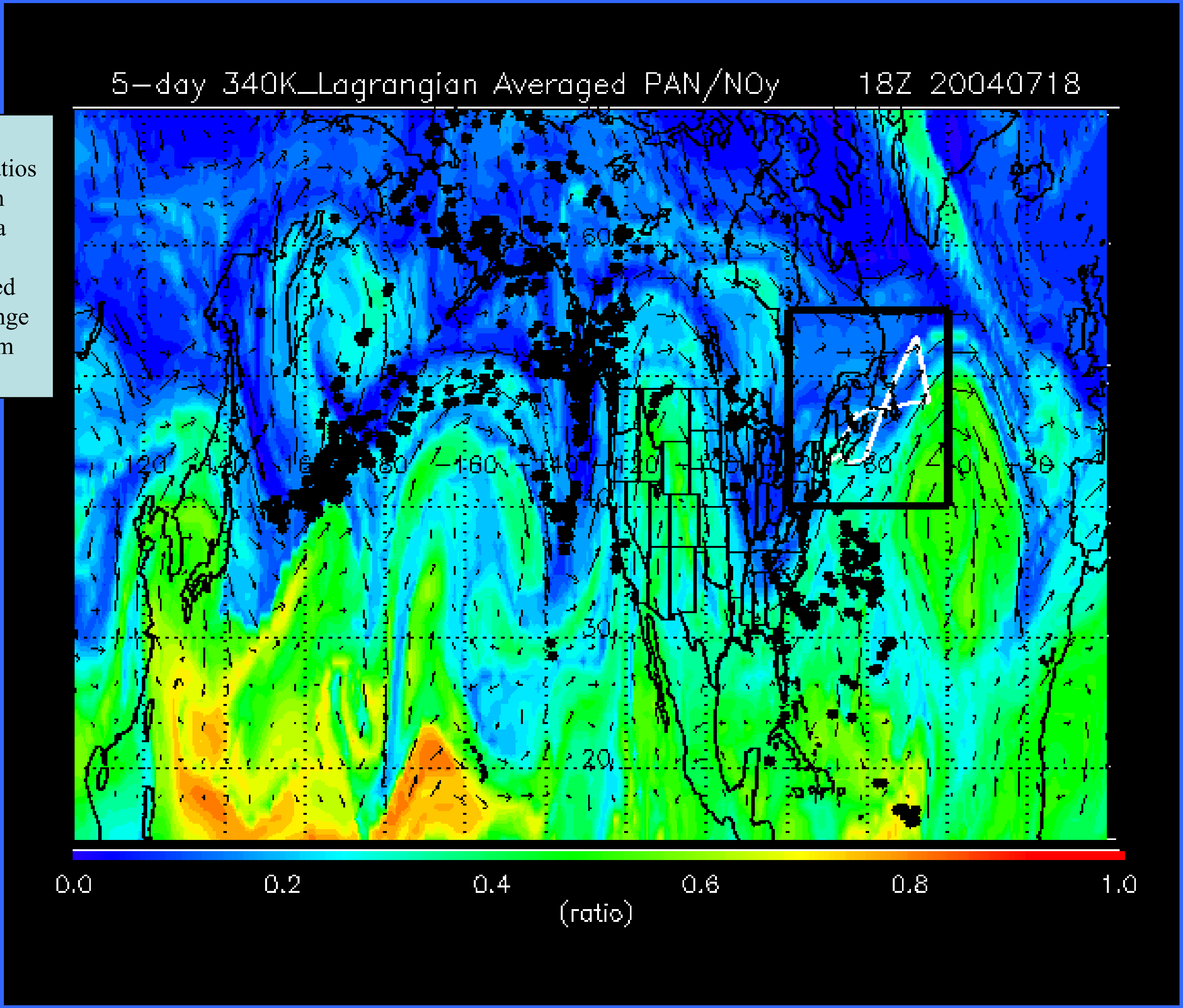
RAQMS$_G$ 340K Lagrangian Analyses 18Z July 18th, 2004



Asian transport to south of stratospheric airmass associated with large Rossby wave breaking event. Stratospherically influenced air on edge of Bermuda high.

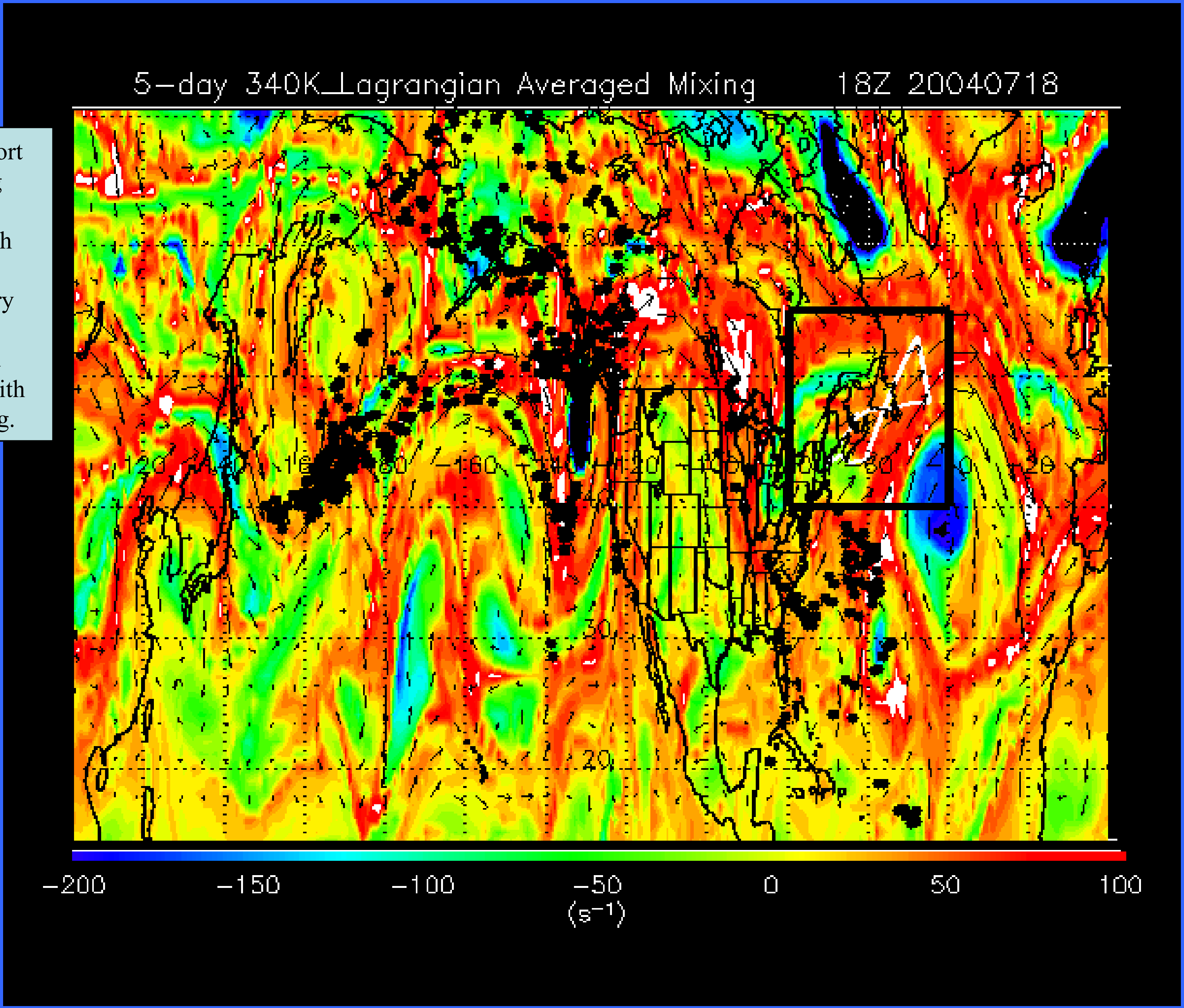
RAQMS$_G$ 340K Lagrangian Analyses 18Z July 18th, 2004

Enhanced PAN/NOy ratios over Western US, Bermuda High, and associated with long-range transport from Asia.



RAQMS$_G$ 340K Lagrangian Analyses 18Z July 18th, 2004

Asian transport within strong mixing zone. Bermuda high shows inner core with very weak mixing (stirring) and outer edge with strong mixing.



A Climatology of Rossby Wave Breaking along the Subtropical Tropopause

GREGORY A. POSTEL AND MATTHEW H. HITCHMAN

Department of Atmospheric and Oceanic Sciences, University of Wisconsin—Madison, Madison, Wisconsin

Rossby Wave Folding events

Rossby Wave breaking is an upstream source for stratospheric ozone in the upper troposphere.

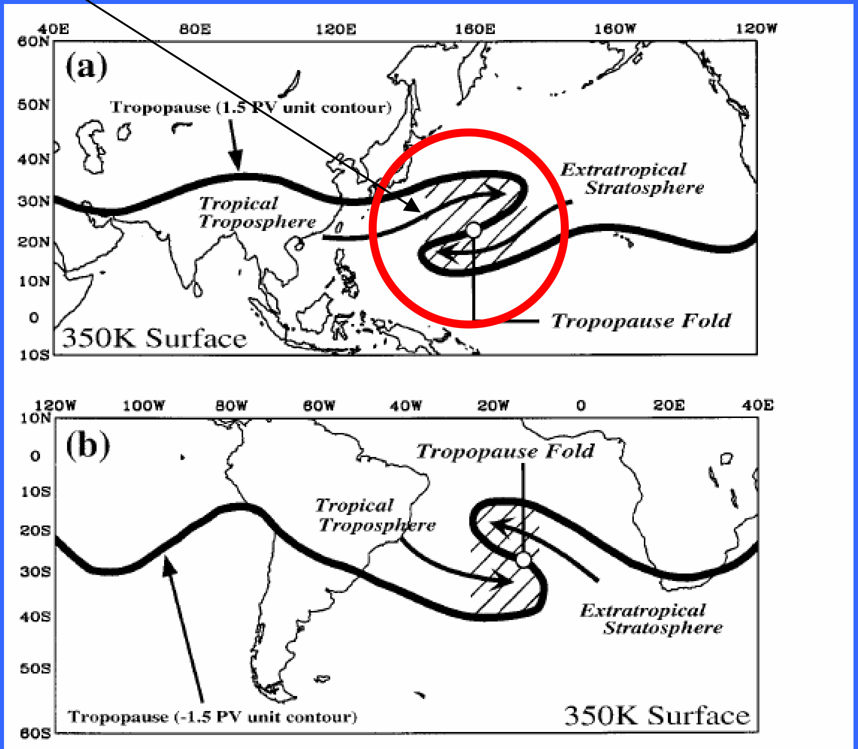


FIG. 1. Schematic of Rossby wave breaking events over (a) the North Pacific and (b) the South Atlantic, on the 350 K isentropic surface. The thick contours represent the tropopause. The hatched regions denote surf zones, where the meridional gradient of PV is regionally reversed.

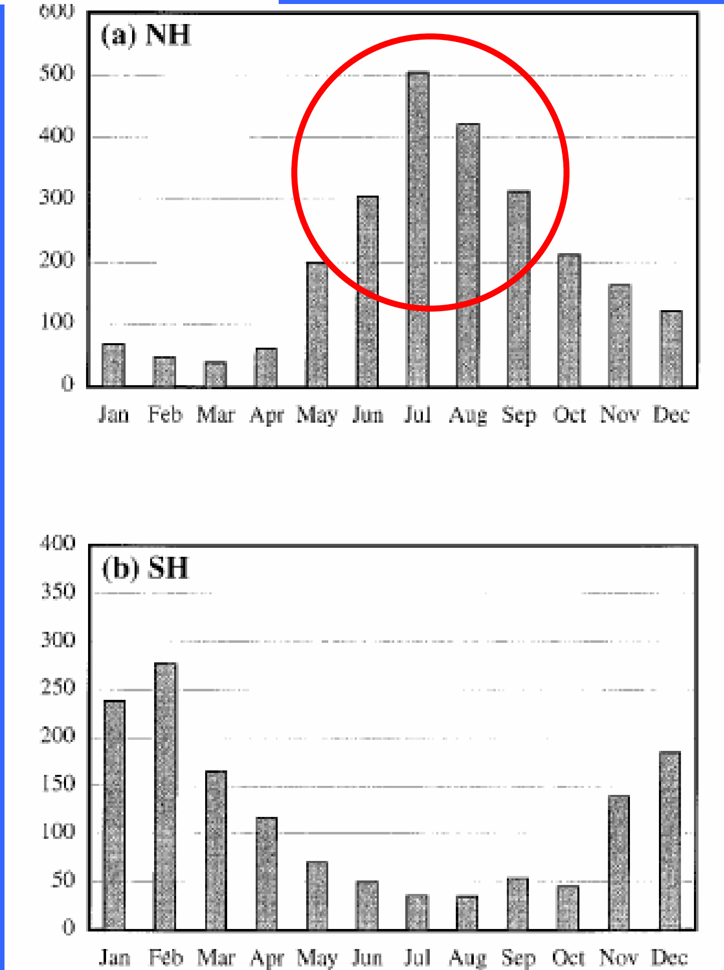


FIG. 3. Histogram of the total number of tropopause folds detected during the 1986–95 period at 350 K, as a function of month, for (a) the NH and (b) the SH.

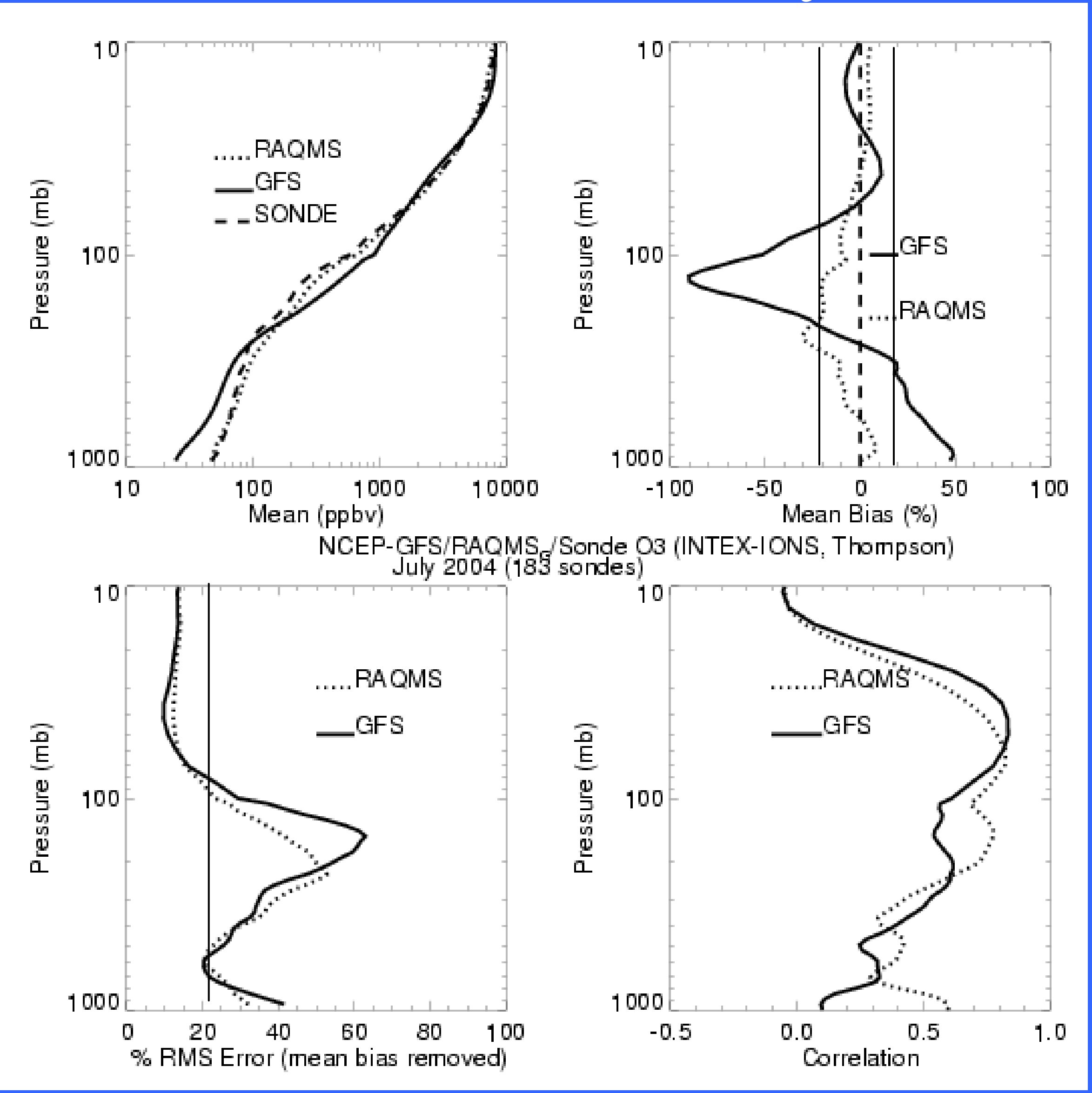
Asian outflow is likely to be highly influenced by STE associated with Rossby wave breaking in Western Pacific during June-August.

Indirect Validation of SAGE III Limb Scattering Measurements: (D. Rault, Lead)

- Through coordination with the SAGE III science team (D. Rault, C. Trepte, NASA/LaRC) , special limb scattering measurements where conducted during May, July and August, 2004 over the Eastern US and North Atlantic in support of the 2004 INTEX-NA mission.

- We have begun indirect validation studies where RAQMS ozone analyses are used as a transfer standard between the INTEX-NA IONS ozonesonde data and contemporaneous, but not coincident, SAGE III LS measurements.

RAQMS and NOAA GFS vs IONS ozonesonde: July 2004

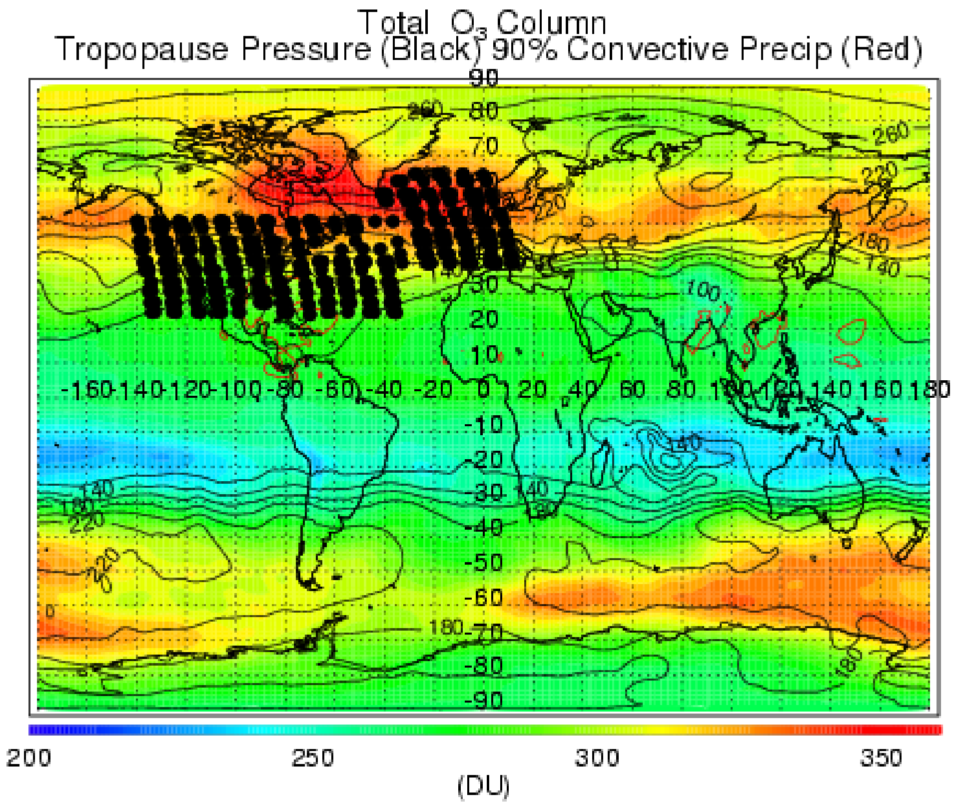


RAQMS O_3 Mean bias* and RMS errors <20% above 100mb.

*Reduction in the high biases relative to GFS due to assimilation of high vertical resolution solar occultation measurements instead of SBUV2.

Reduction in tropospheric low biases relative to GFS due to the inclusion of realistic tropospheric ozone photochemistry.

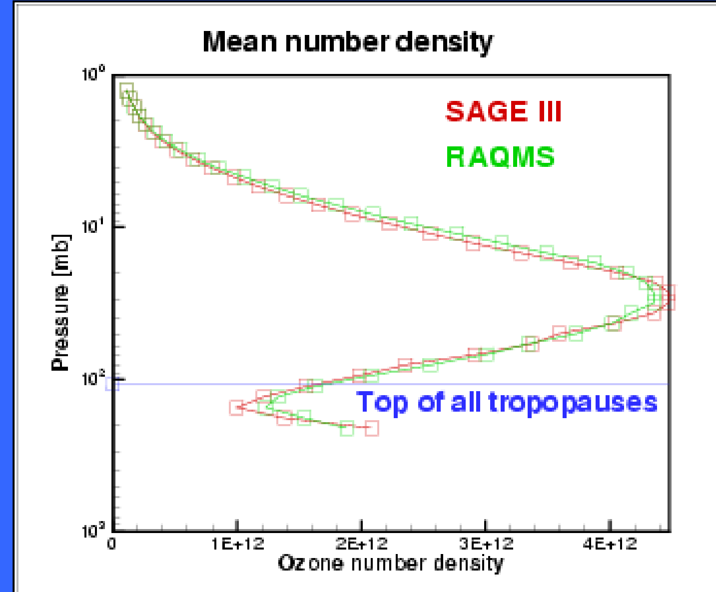
RAQMS vs SAGE III Limb Scattering (Rault): July 2004

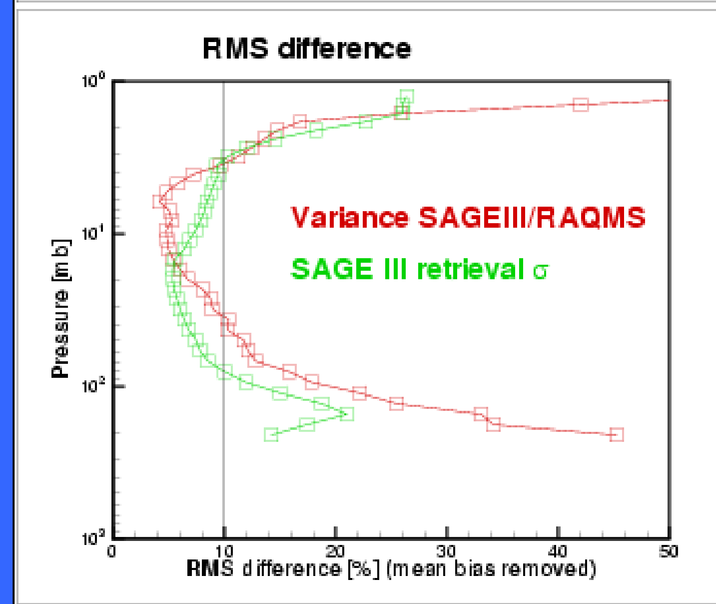


RAQMS Total Column Ozone July 2004.

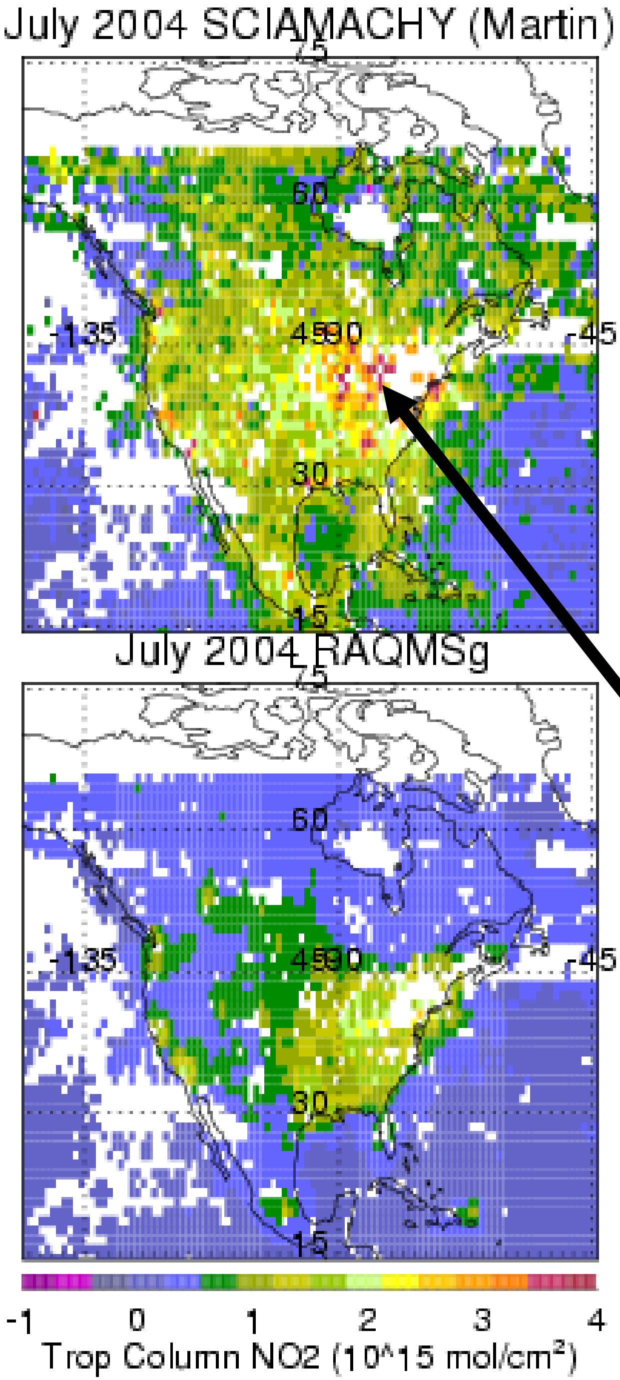
RAQMS vs SAGE III LS O_3 Mean Bias and RMS Errors are </= Sonde statistics.

RMS Error is consistent with estimated SAGE III LS retrieval uncertainty.

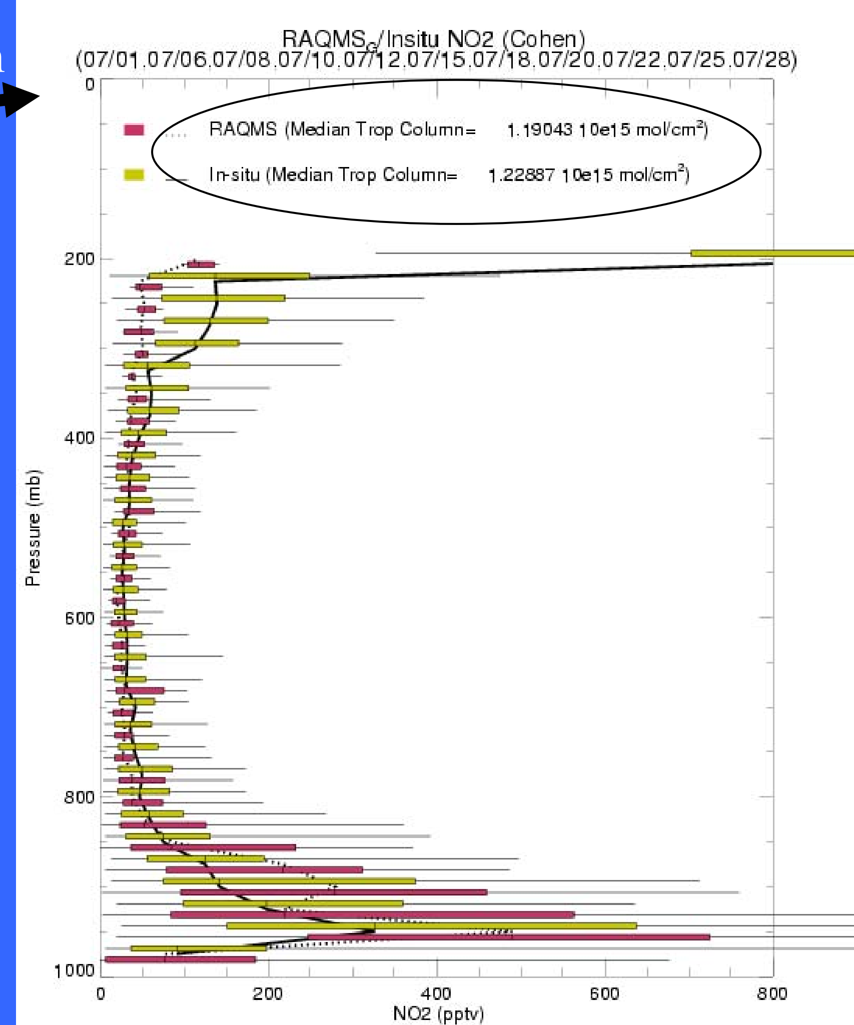




Indirect validation of SCIAMACHY (Martin) tropospheric NO2 column



RAQMS Median Column compares very well with insitu estimates within domain sampled by DC8 (Eastern US).

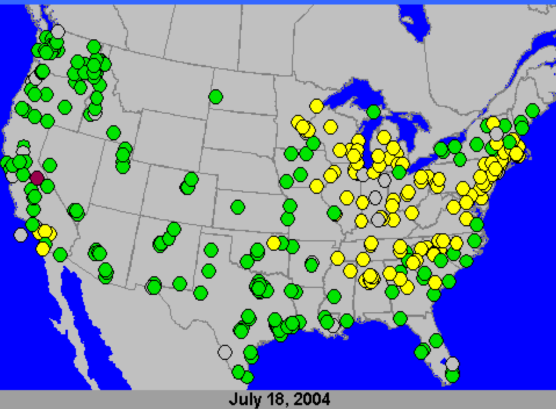


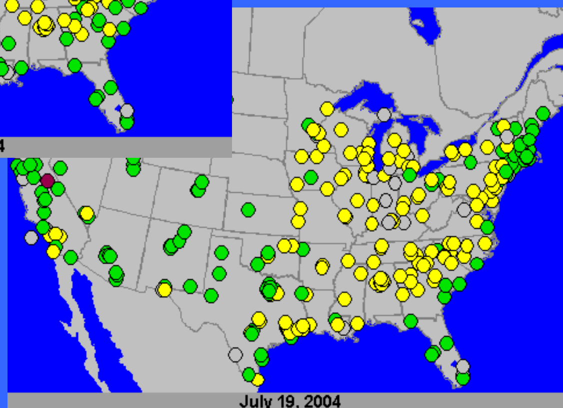
RAQMS Mean column is generally low relative to SCIAMACHY. Particularly in urban centers and generally over the western US.

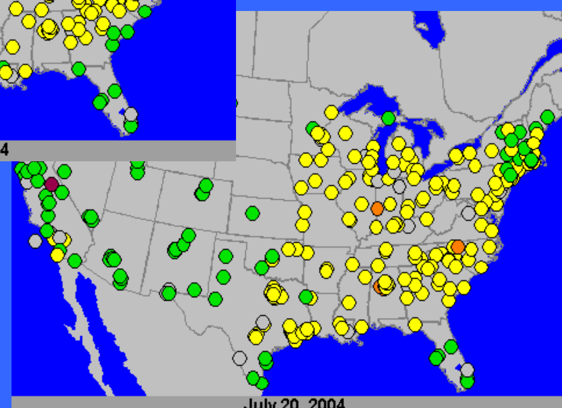
RAQMS Regional Aerosol Assimilation and Forecasting Case Study:
(C. Kittaka, Lead)

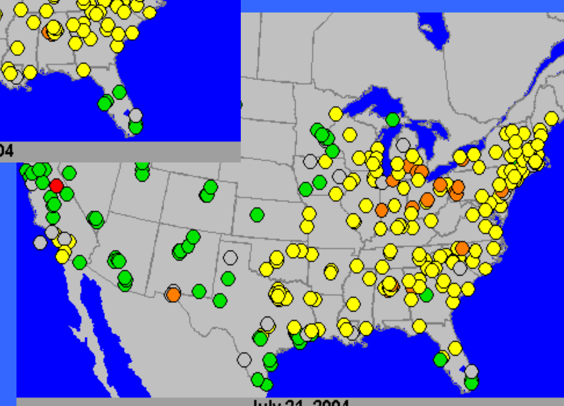
July 18-July 22, 2004 PM2.5 AQI event

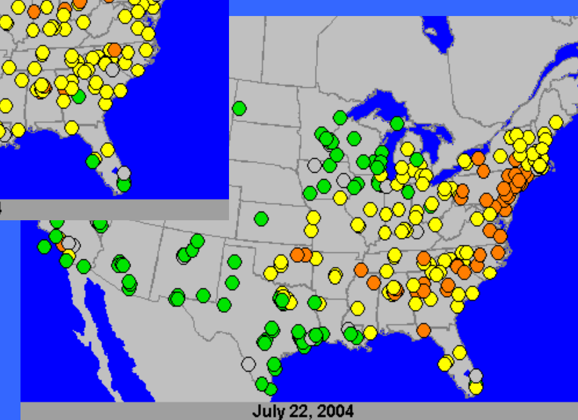
EPA AIRNow PM25 AQI:
July 18- July 22, 2004

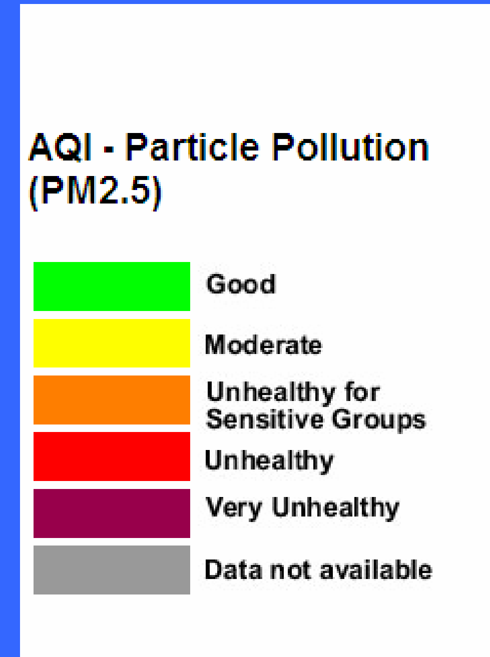








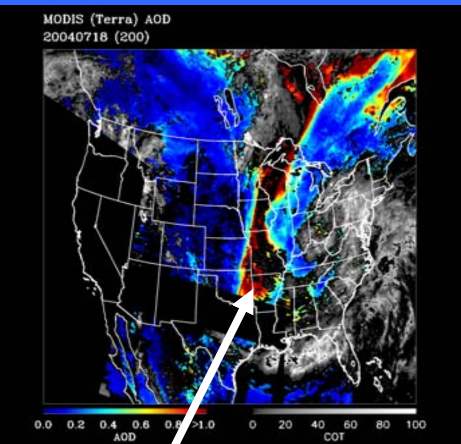




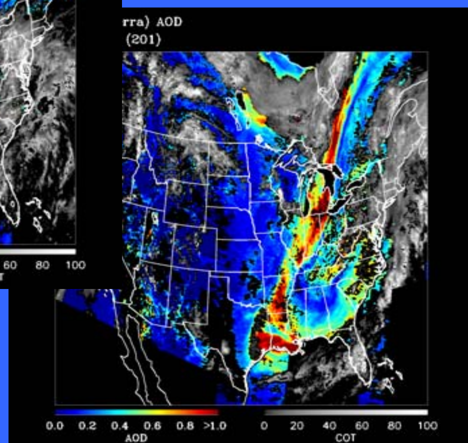
Moderate PM2.5 AQI on July 20, Unhealthy for sensitive groups in Great Lakes on 21st then SE-NE by July 22, 2004.

MODIS Aerosol Optical Depth (A. Chu): July 18- July 22, 2004

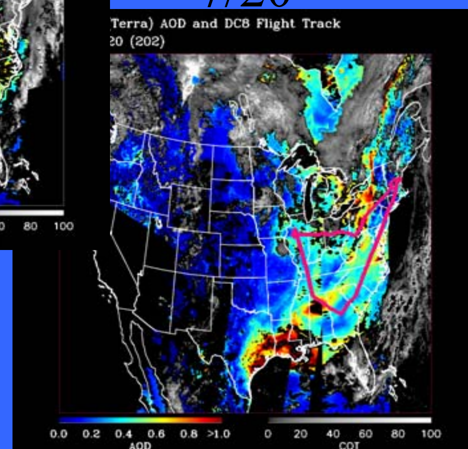
7/18



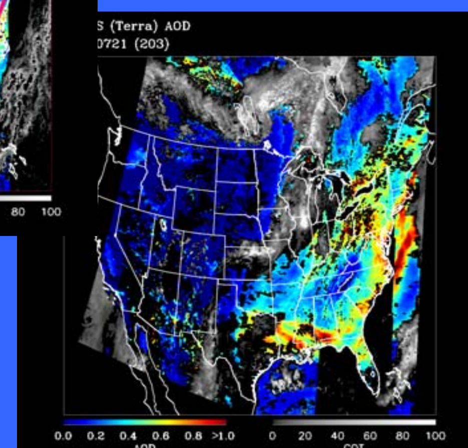
7/19



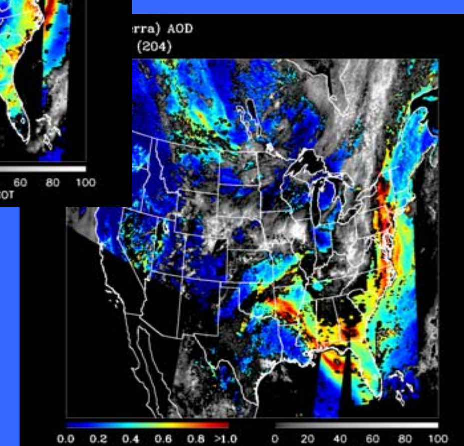
7/20



7/21



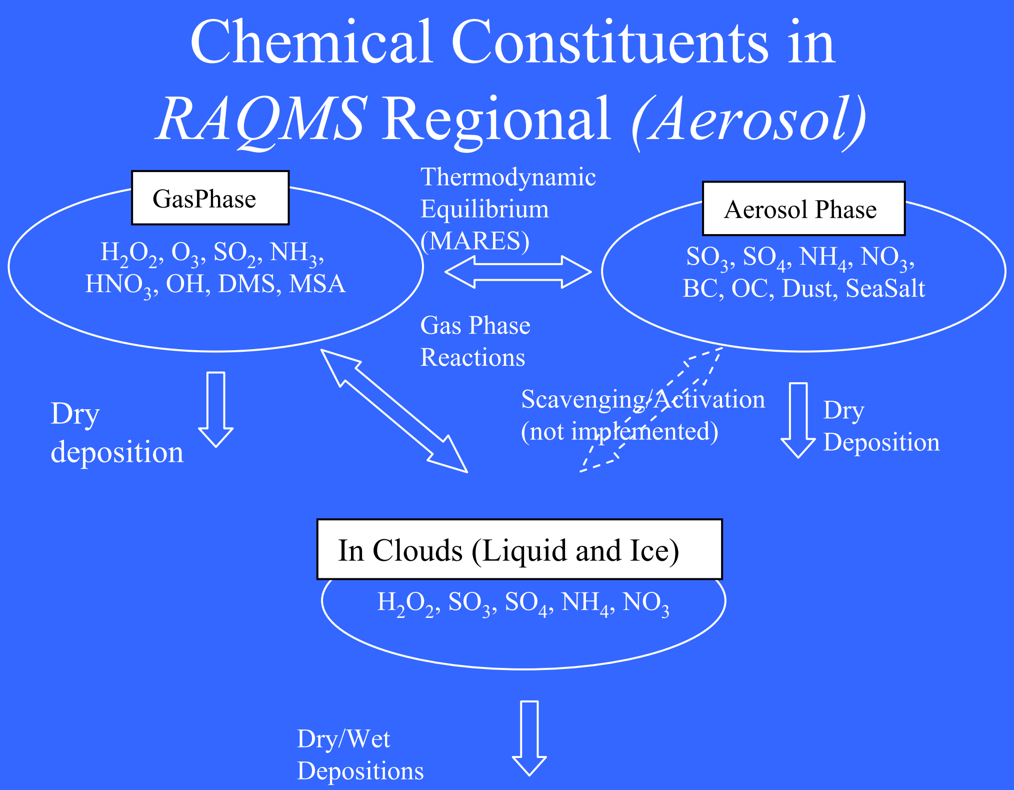
7/22



Smoke from Alaskan Forest Fires

Link between Alaskan smoke and US AQ?

RAQMS regional Aerosol Forecast MODIS AOD assimilation

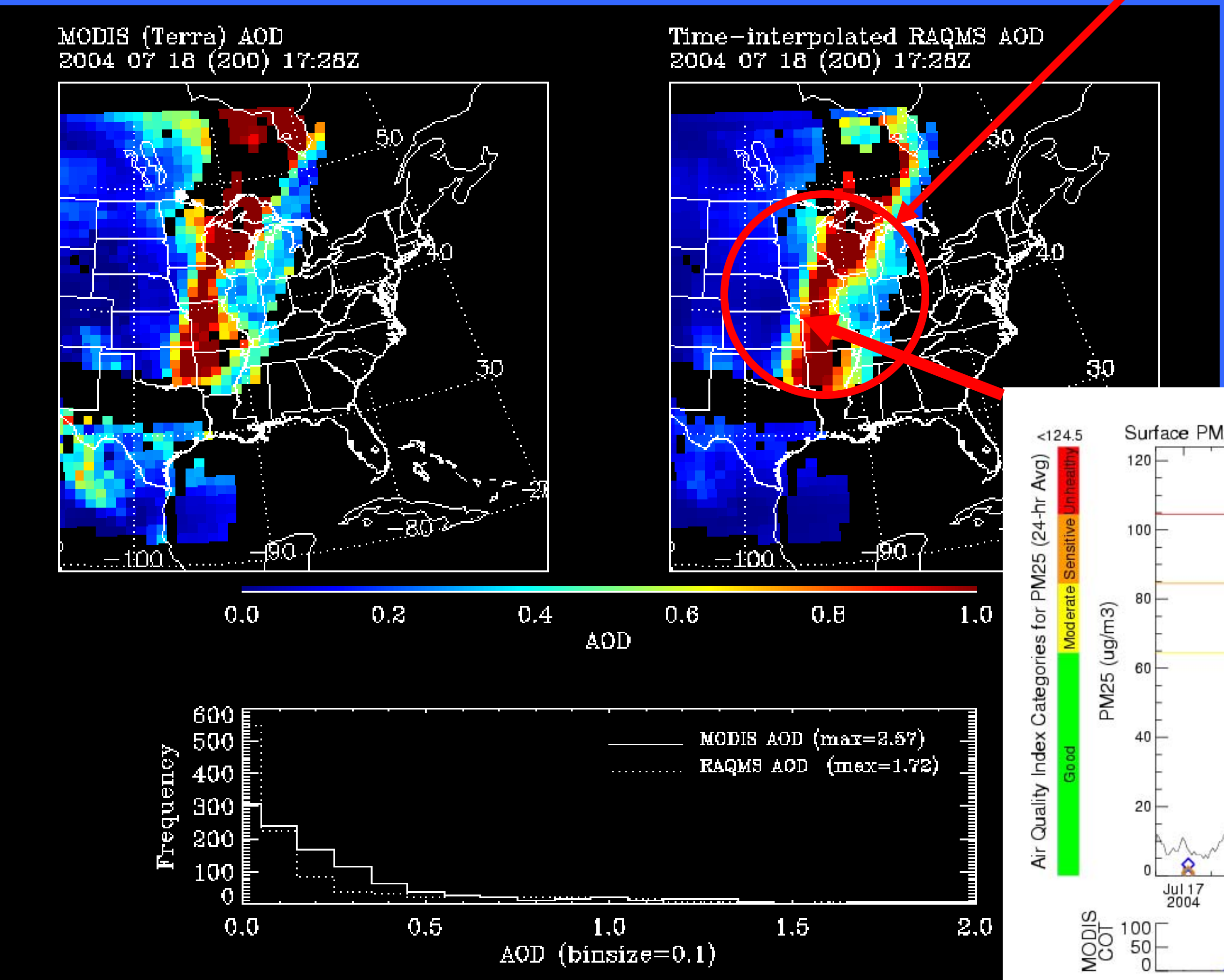


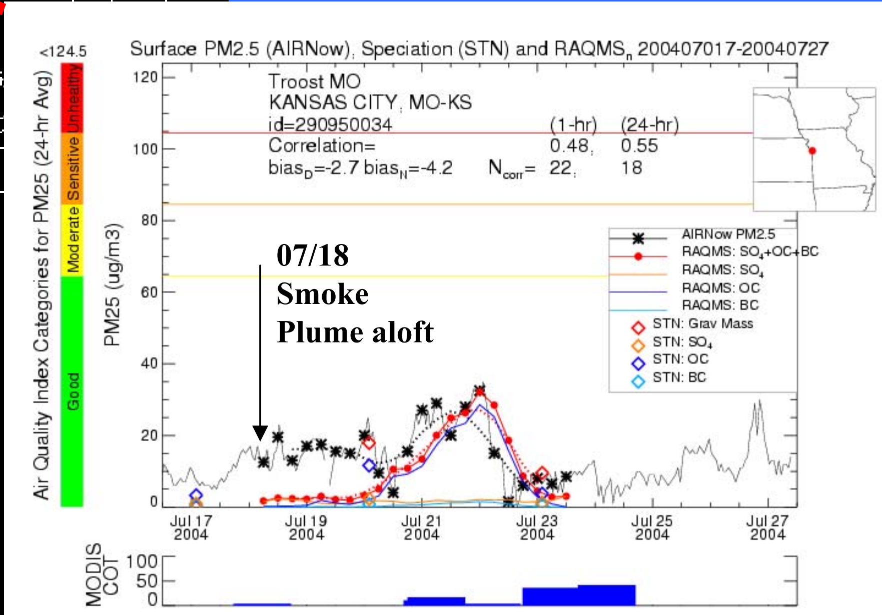
Does Smoke from Alaskan Fires get entrained within CONUS boundary Layer?

- **Initialized on July 15, 2004**
- **UWNMS Dynamical Core**
- **80Km Continental US/EDAS Met BC/IC**
- **$RAQMS_G$ Chemical BC/IC**
- **GOCART background climatological IC/BC**
- **Sulfate [Kittaka, 2004], Dust, Sea Salt, Carbonaneous Aerosol from GOCART [provided by Mian Chin, GSFC]**
- **Nitrate and Ammonium from GOES-CHEM [provided by Rokjin Park, Harvard]**
- **BC+OC perturbations added above BL on 18Z July 18, 2004, vertical extent constrained by SSEC Lidar**
- **One MODIS AOD assimilation cycle used for final constraint on total AOD**

MODIS vs RAQMS AOD
17:28Z July 18, 2004

Assimilated Smoke plume



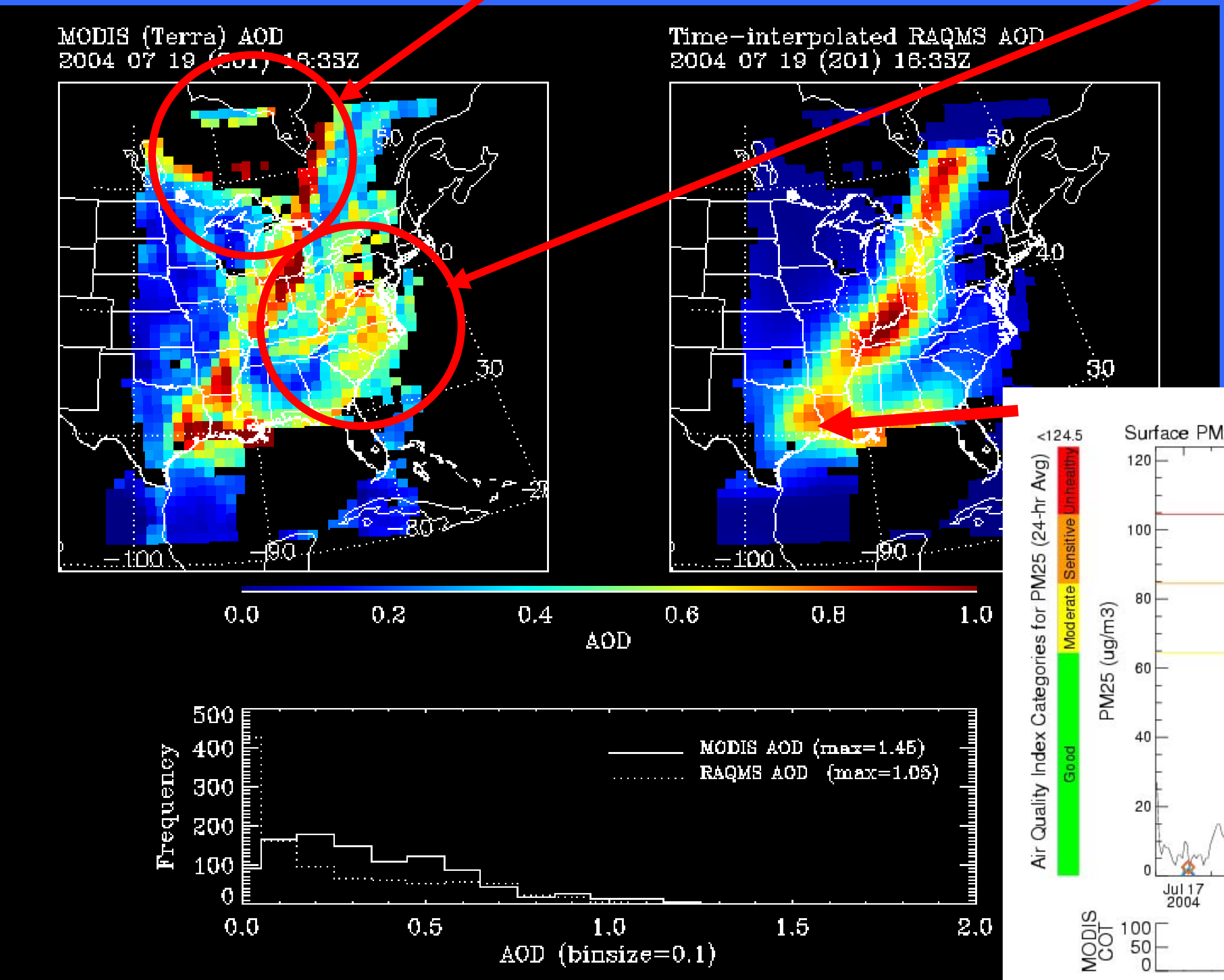


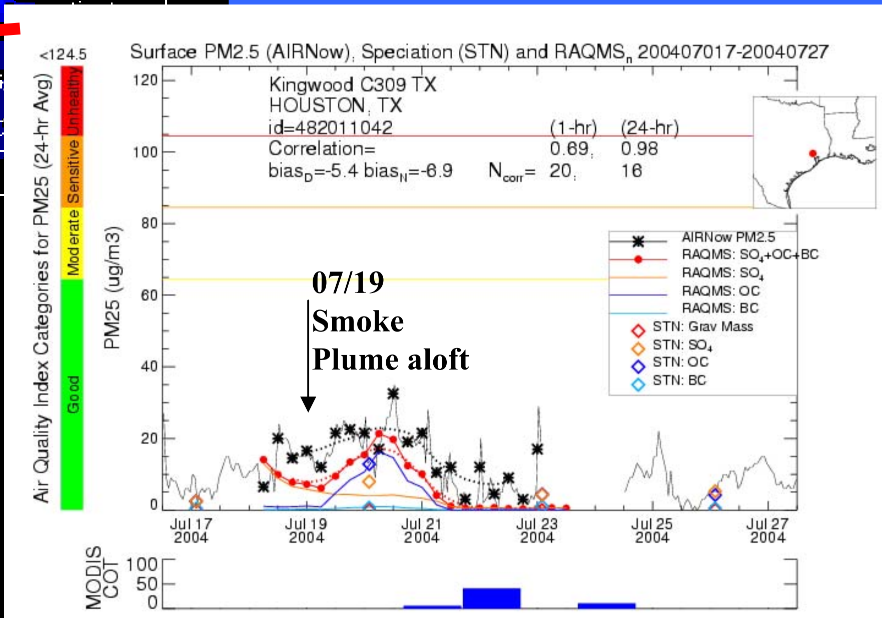
MODIS vs RAQMS AOD 16:33Z July 19, 2004

New Smoke plume
(not assimilated)

Sulfate plume previously obscured by clouds
(not assimilated)

RAQMS/MODIS AOD show good agreement elsewhere

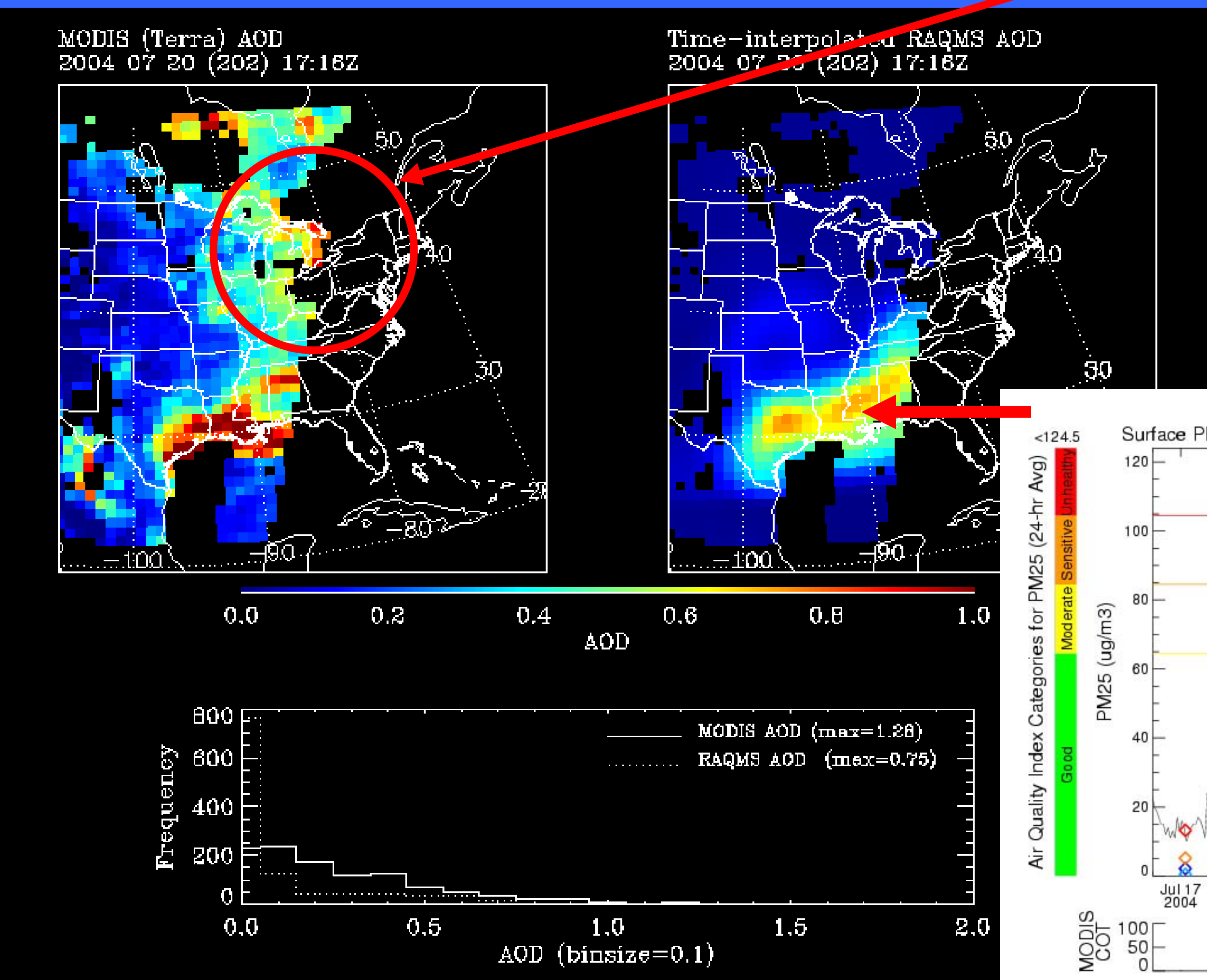


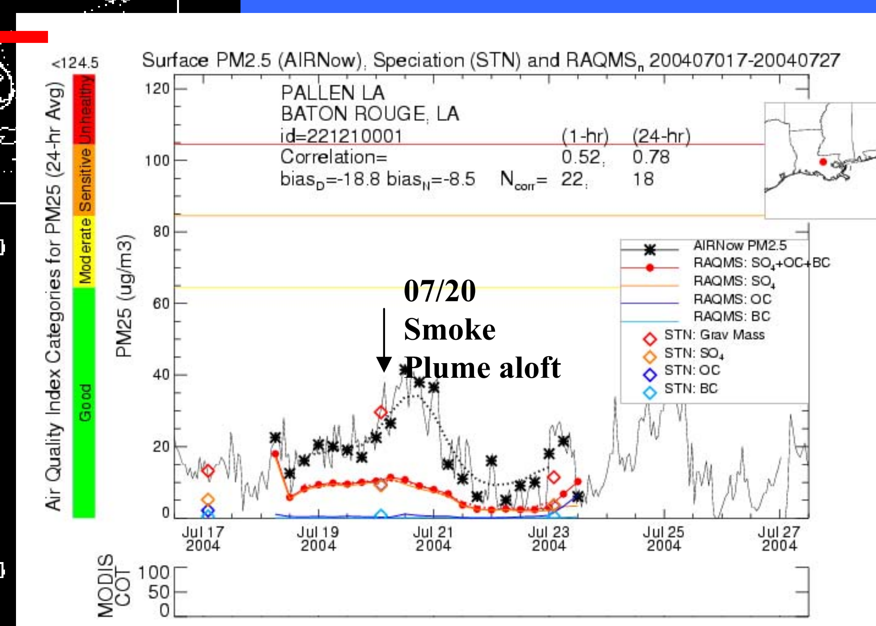


MODIS vs RAQMS AOD
17:16Z July 20, 2004

New Smoke plume transported Southeastward

RAQMS underestimates Gulf Coast AOD

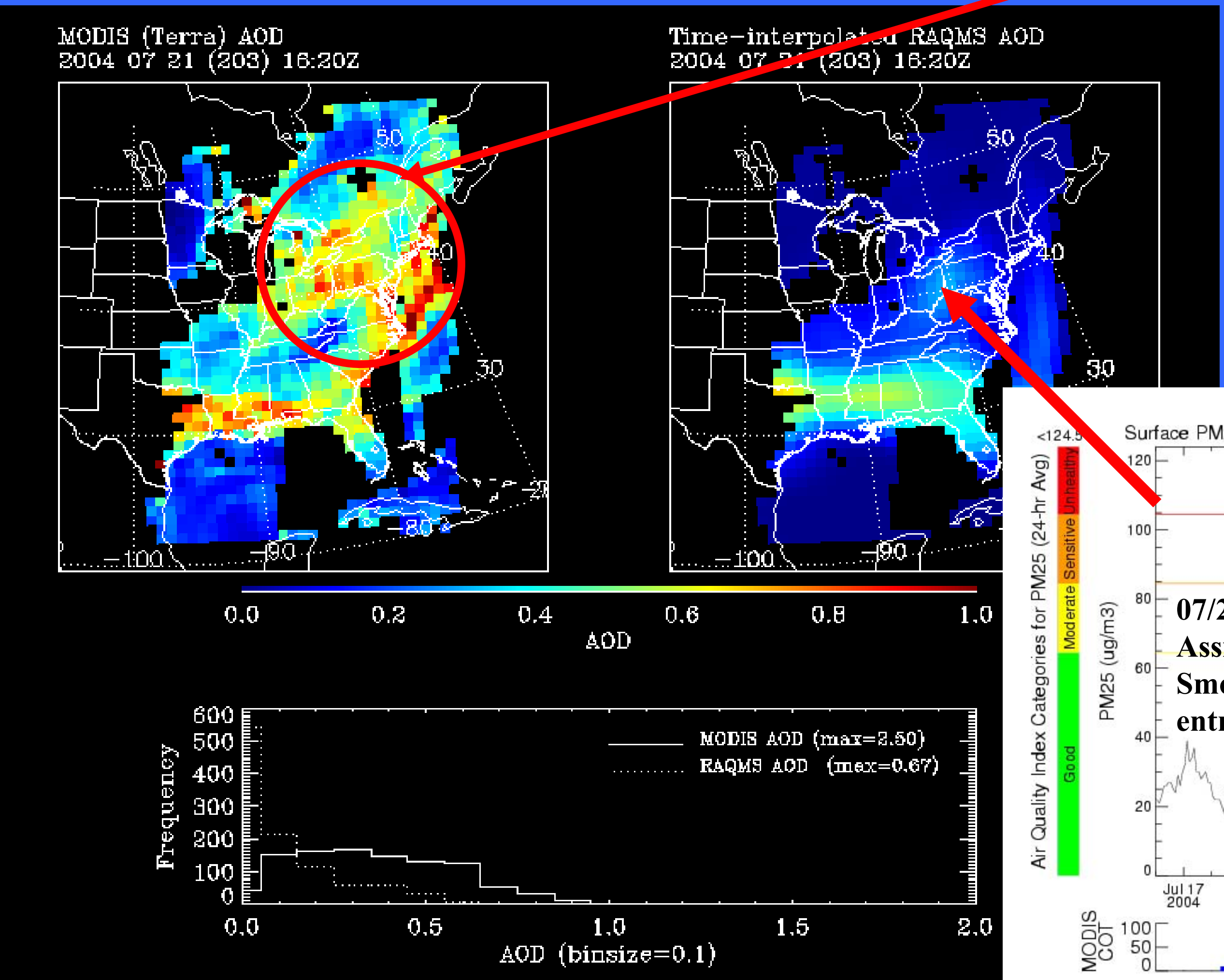


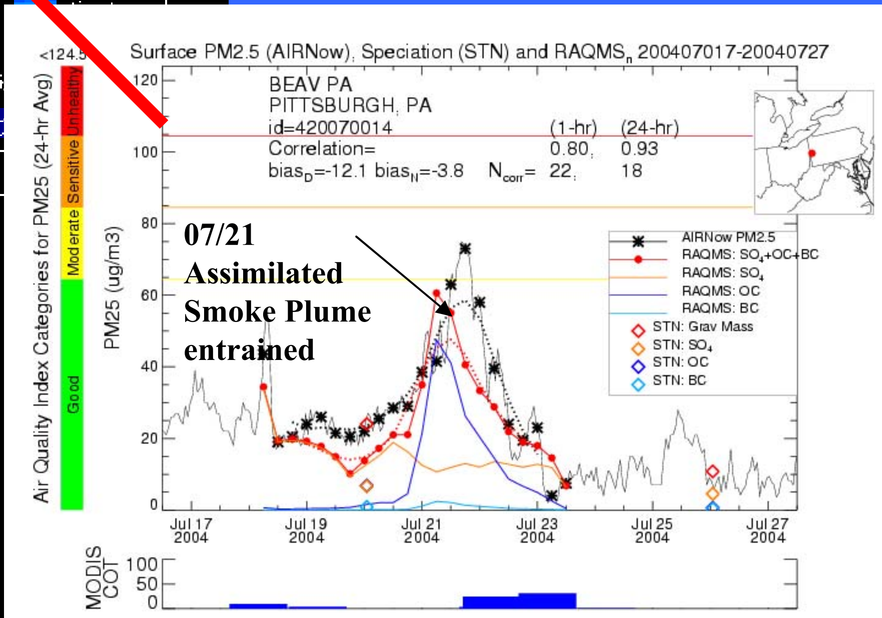


MODIS vs RAQMS AOD 16:20Z July 21, 2004

New Smoke and Sulfate plume move Northeastward

RAQMS significantly underestimates PA AOD, but agrees will with EPA PM2.5 at surface → New Smoke aloft

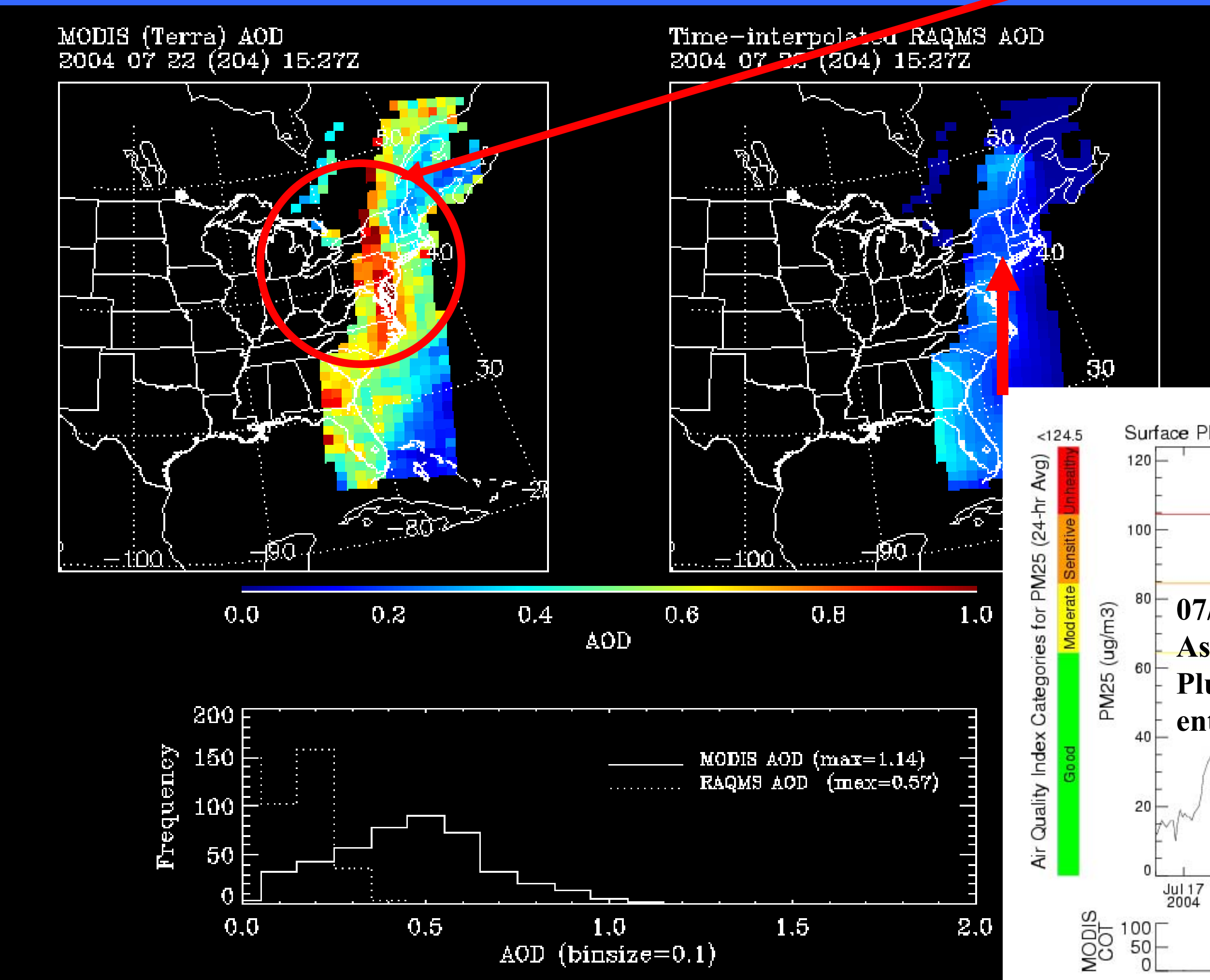


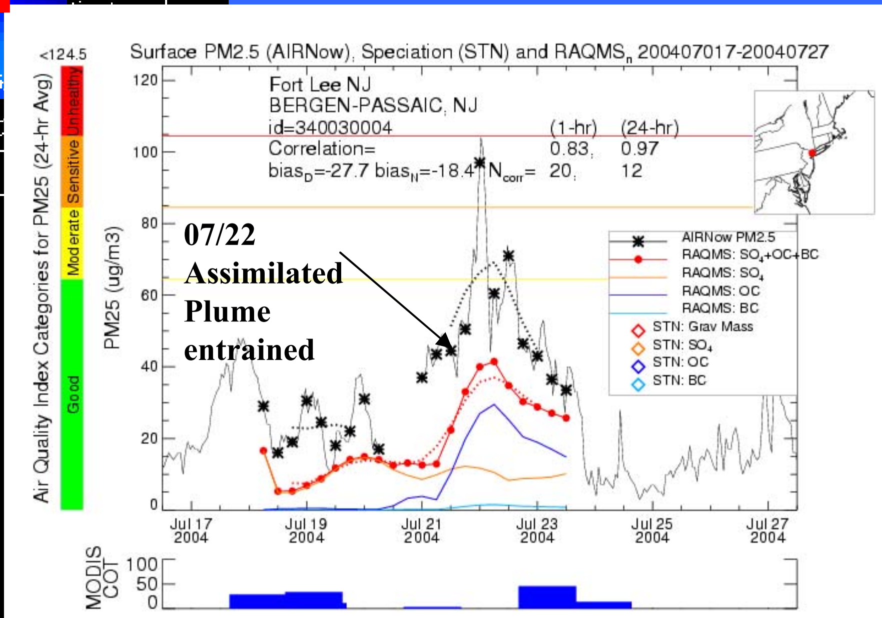


MODIS vs RAQMS AOD
15:27Z July 22, 2004

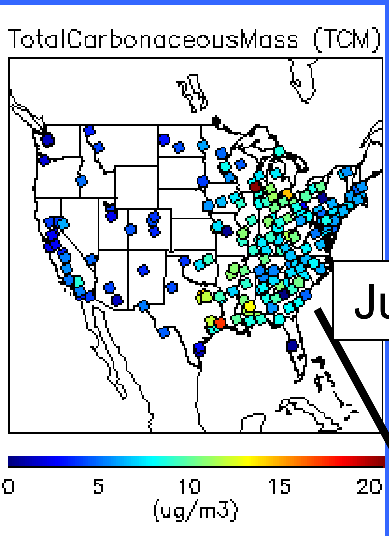
New Smoke and
Sulfate plume merge

AOD over Eastern
Seaboard is significantly
underestimated, PM2.5 is
Underpredicted by ~30%

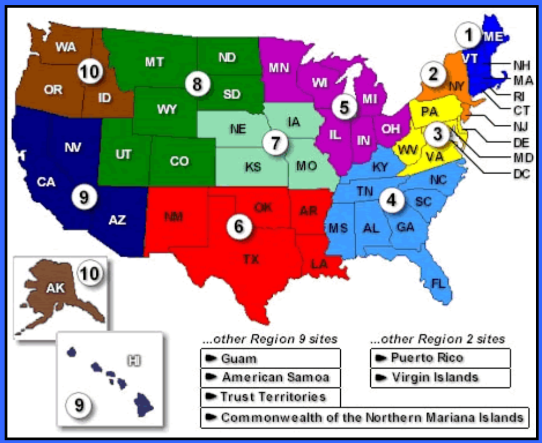


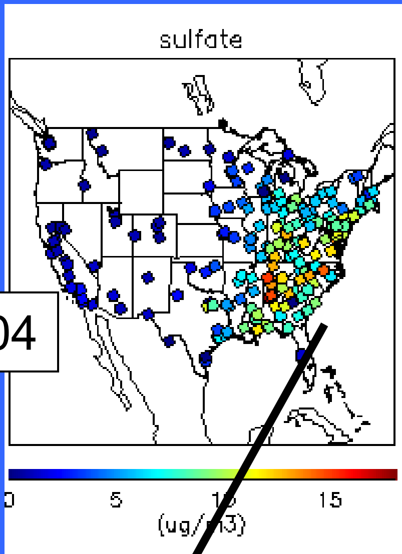


EPA Surface Speciation Data July 2004

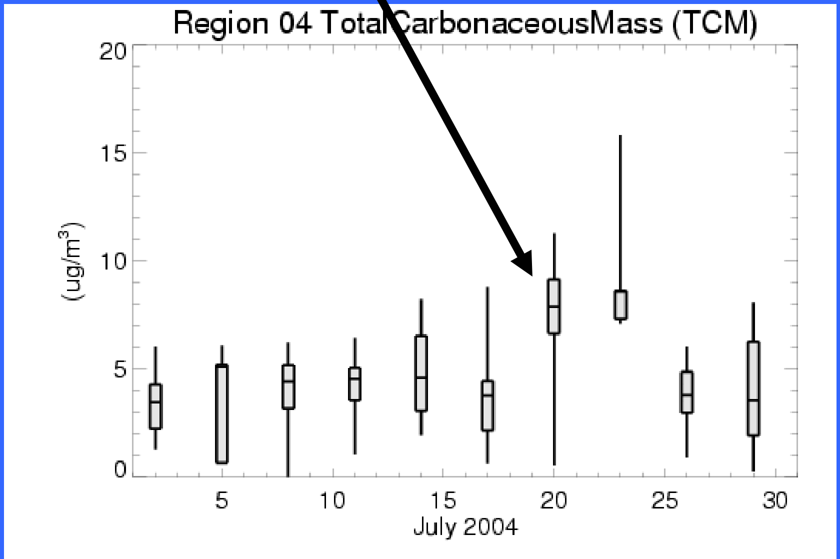


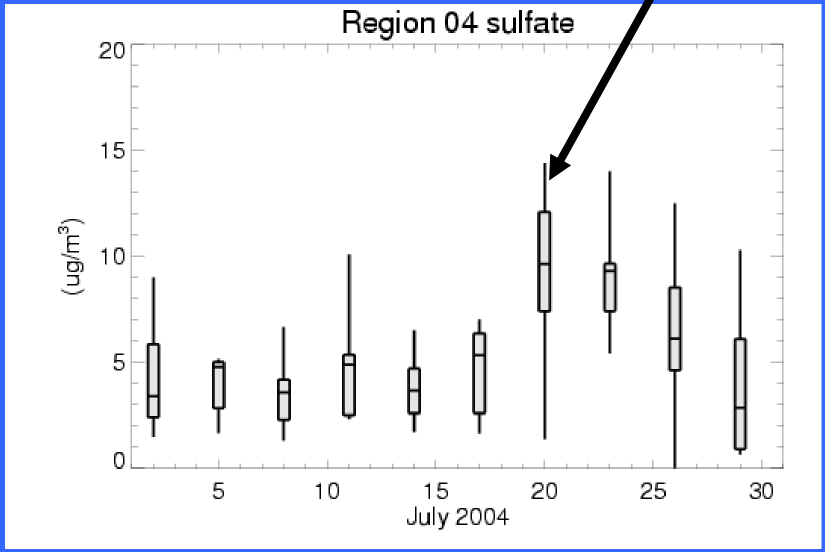
July 20, 2004





July 20, 2004





PM2.5 AQI over the SE US (Region 04) was influenced by both local sulfate and Alaskan carbonaceous aerosols during July 22 AQI event.

Continuing Research/Manuscript Plans:

Alaskan Fires:

•Regridded (based on daily MODIS fire counts) climatological Alaskan/Canadian emission runs currently being conducted with $RAQMS_G$ (problems with plume transport within polar filter).

•Development of NRT fire emissions based on MODIS fire counts, climatological carbon load and Heines index based fire intensity estimates (with Amber Soja, NRC/LaRC).

Assimilation:

•$RAQMS_G$ SAGE III Limb Scattering impact studies in preparation for INTEX-B OMI+SAGE III Limb Scattering Assimilation

•$RAQMS_N$ MODIS AOD Assimilation in preparation for CALIPSO launch.

Regional budgets

•Conduct Continental US NOy and O3 budget studies (similar to RAQMS SE Asian O3 budget during TRACEP)

• Conduct $RAQMS_N$ two-scale STE analyses within Pacific Rossby wave breaking events.

Lagrangian Analysis

•Statistical analysis of Lagrangian maps to determine dominate source/receptor relationships and magnitudes of chemical transformation during long-range transport.

Extra Figures

RAQMS unified (strat/trop) chemistry

(55 species/families explicitly transported, 86 calculated, PCE assumptions for "fast" species)

1) Ox
2) Noy
3) Cly
4) Bry
5) HNO3
6) N2O5
7) H2O2
8) HCl
9) ClONO2
10) OClO
11) N2O
12) CFCl3 (F11)
13) CF2Cl2 (F12)
14) CCl4
15) CH3Cl
16) CH3CCl3 (MTCFM)
17) CH3Br
18) CF3Br (H1301)

19) CF2ClBr (H1211)
20) HF
21) CFClO
22) CF2O
23) CH4
24) HNO4
25) HOCl
26) H2O
27) NO3
28) NO2
29) CH2O
30) CH3OOH
31) CO
32) HBr
33) BrONO2
34) HOBr
35) BrCl
36) Cl2

Stratosphere+CH4&CO oxidation

37) C2H6 (ethane, 2C)
38) ALD2 (acetaldehyde+higher group, 2C)
39) ETHOOH (ethyl hydrogen peroxide, 2C)
40) PAN (2C)
41) PAR (paraffin carbon bond group, 1C)
42) ONIT (organic nitrate group, 1C)
43) AONE (acetone, 3C)
44) ROOH (C3+hydrogen peroxides group, 1C)
45) MGLY (methylglyoxal, 3C)
46) ETH (ethene, 2C)
47) XOLET (terminal olefin carbon group, 2C)
48) XOLEI (internal olefin carbon group, 2C)
49) XISOP (isoprene, 5C)
50) XISOPRD (isoprene oxidation product-long lived, 5C)
51) PROP_PAR (propane paraffin, 1C)
52) CH3OH (methanol)
53) XMVK (methyl vinyl ketone, 4C)
54) XMACR (methacrolein, 4C)
55) XMPAN (peroxymethacryloyl nitrate, 4C)

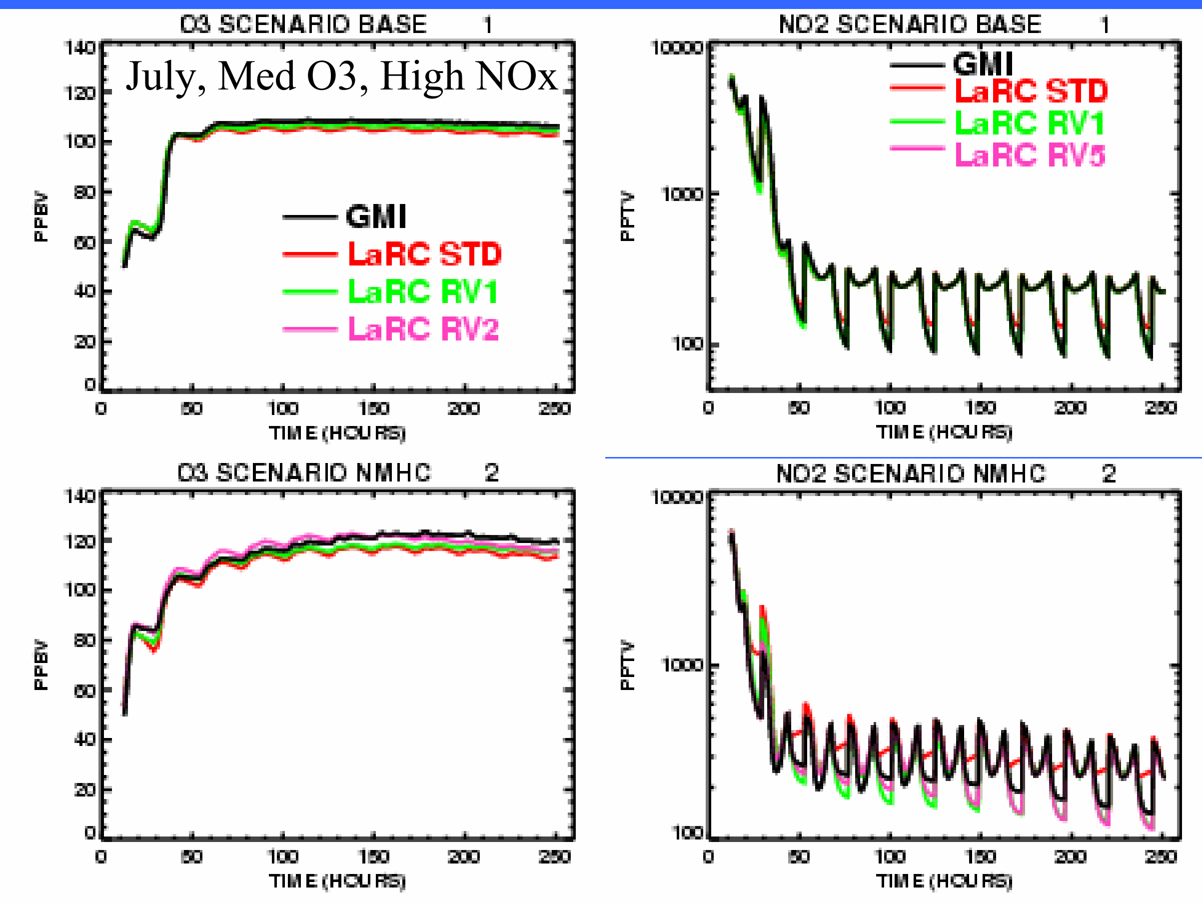
NMHC Chemistry

Chemical families

Ox=O(1D)+O(3P)+O3+NO2+HNO3+2(NO3)+3(N2O5)+HNO4+PAN+MPAN
NOy=N+NO+NO2+NO3+2(N2O5)+HNO3+HNO4+BrNO3+ClNO3+PAN+ONIT+MPAN
Cly=HCl+ClONO2+ClO+2(Cl2O2)+OClO+ClO2+2(Cl2)+BrCl+HOCl+Cl
Bry=HBr+BrONO2+BrO+BrCl+HOBr+Br

RAQMS NMHC Treatment

- Explicit treatment of C_2H_6 (ethane), C_2H_4 (ethene) and CH_3OH (methanol) oxidation [Sander et al., 2003]. C_3H_8 (propane) is handled semi-explicitly.
- C_4 and larger alkanes and C_3 and larger alkenes are lumped via a carbon-bond approach [Zaveri and Peters, 1999] which accounts for long-lived species and their intermediates based on the Carbon Bond Mechanism IV [Gery et al., 1989].
- Isoprene is modeled after the Carter 4-product mechanism as modified for RADM2.



10-day diurnal equilibrium runs with/without NMHC conducted as part of the NASA Global Modeling Initiative (GMI) unified chemistry development.

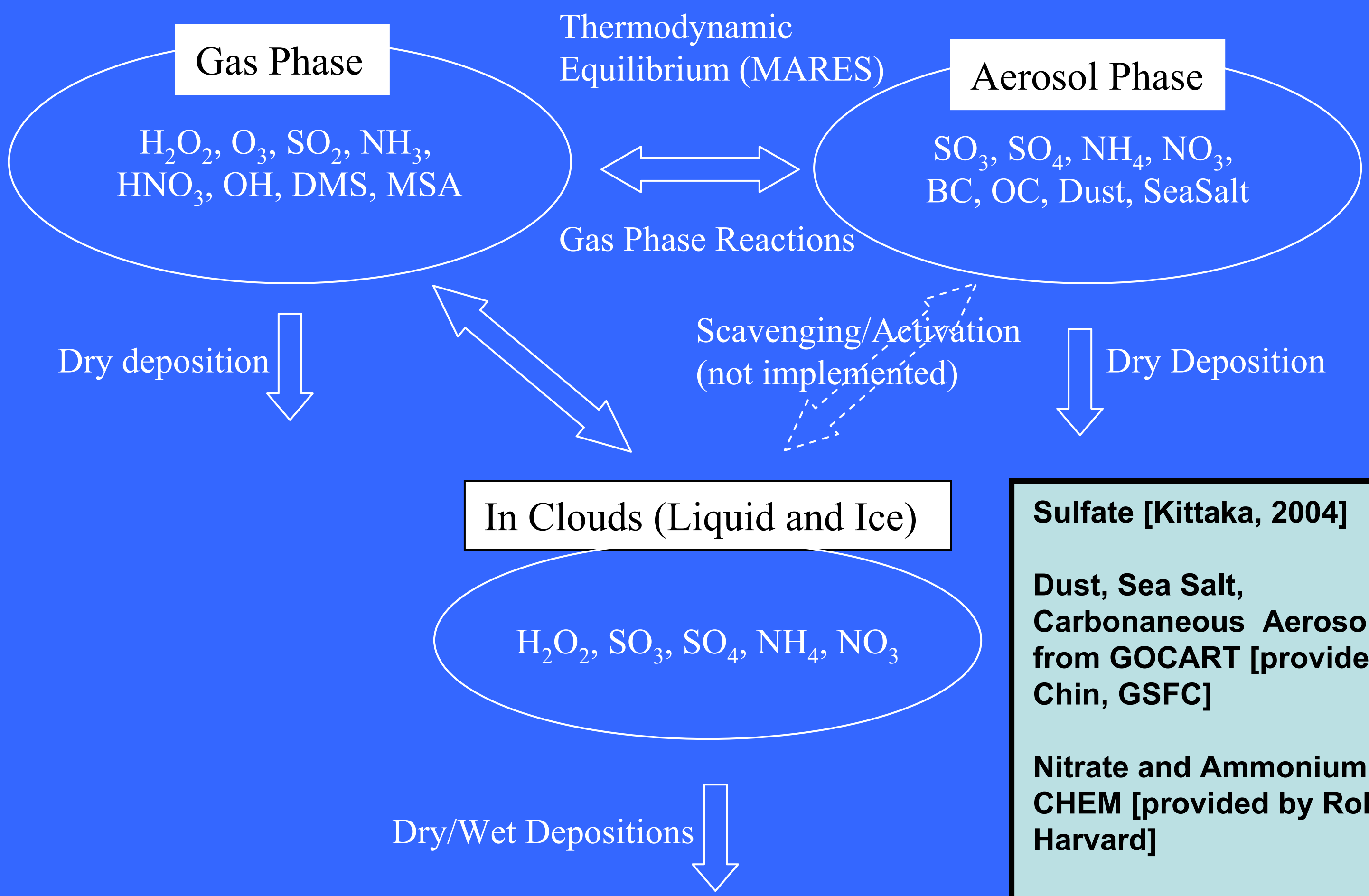
GMI

Harvard mechanism [Bey et al., 2001] with Gear solver for 80 species (all transported in GMI)

LaRC Run Versions

- Standard
- Revised 1: Remove NO3 + peroxy radical rxns
- Revised 2: Revised 1 + ...
  - Peroxide oxidation branching matched to GMI
  - Organic nitrate production matched to GMI
  - RO2 + NO branching matched to GMI

Chemical Constituents in *RAQMS* Regional *(Aerosol)*



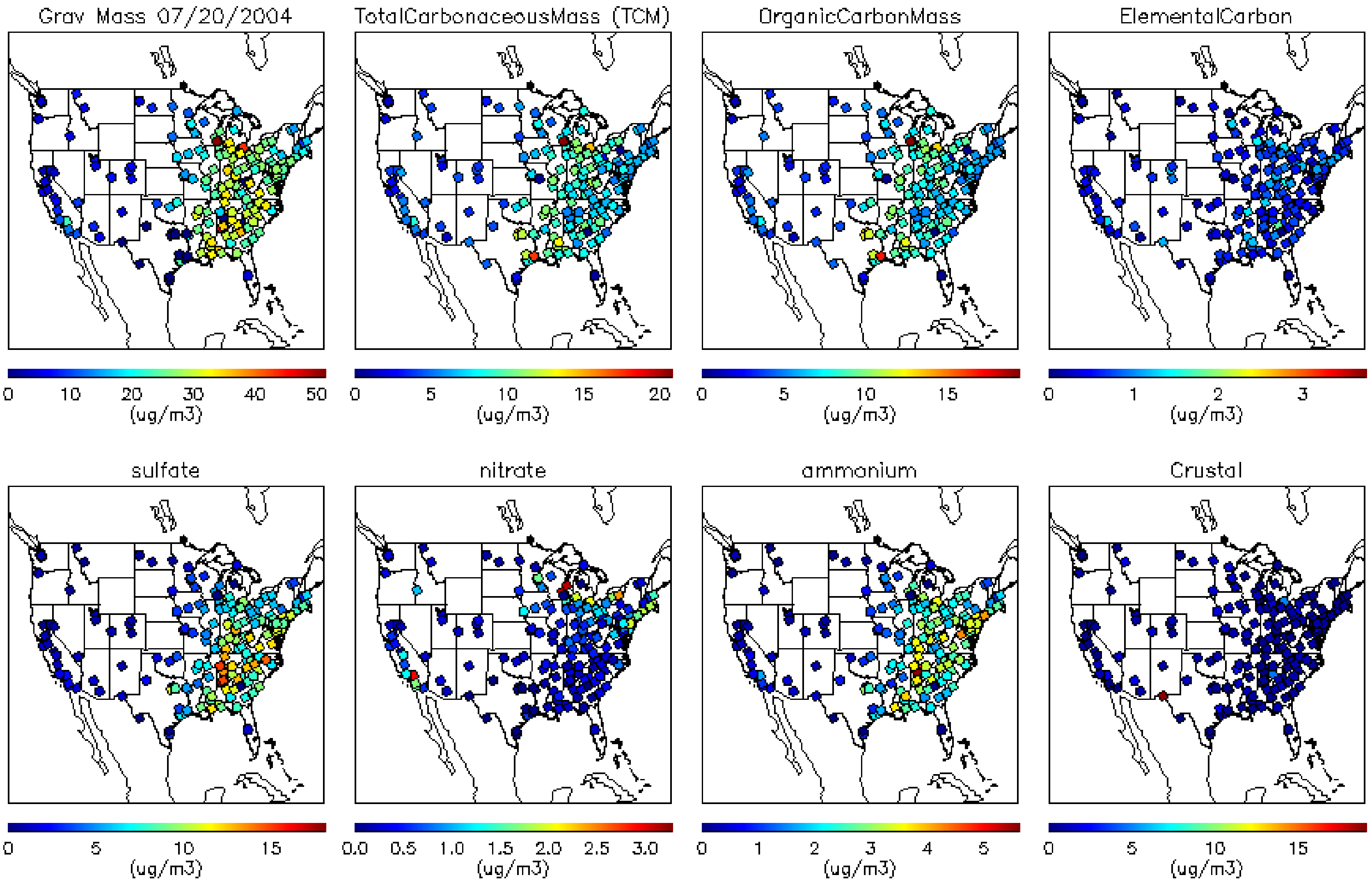
Sulfate [Kittaka, 2004]

Dust, Sea Salt, Carbonaneous Aerosol from GOCART [provided by Mian Chin, GSFC]

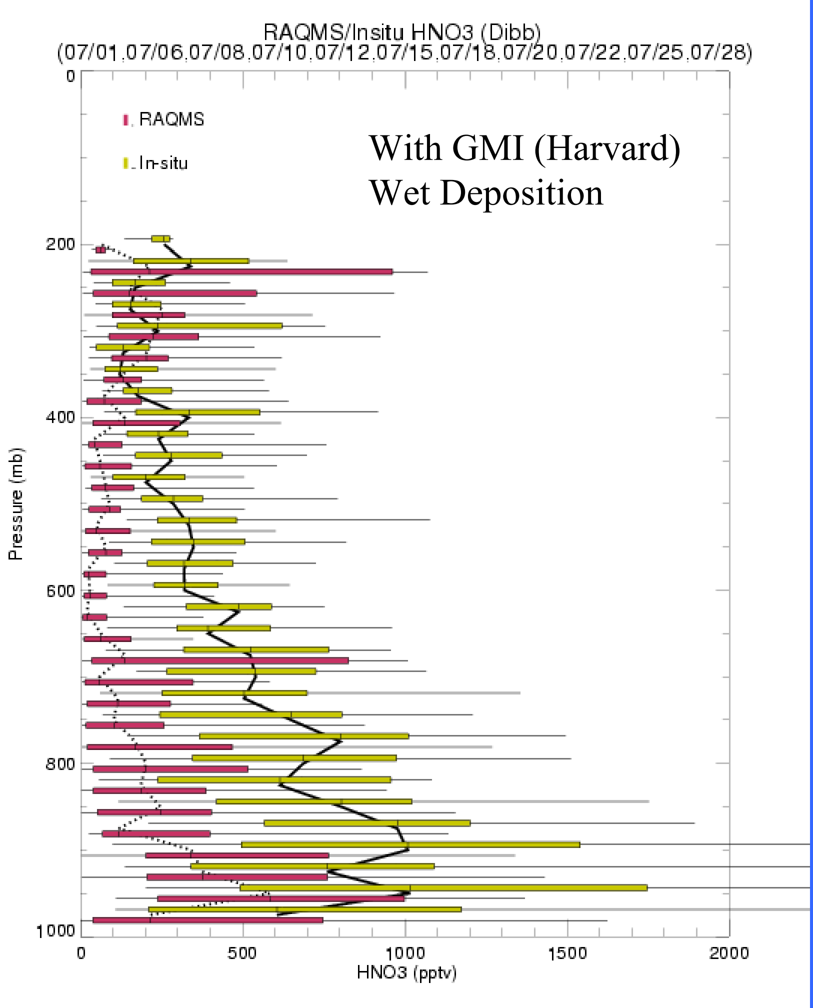
Nitrate and Ammonium from GOES-CHEM [provided by Rokjin Park, Harvard]

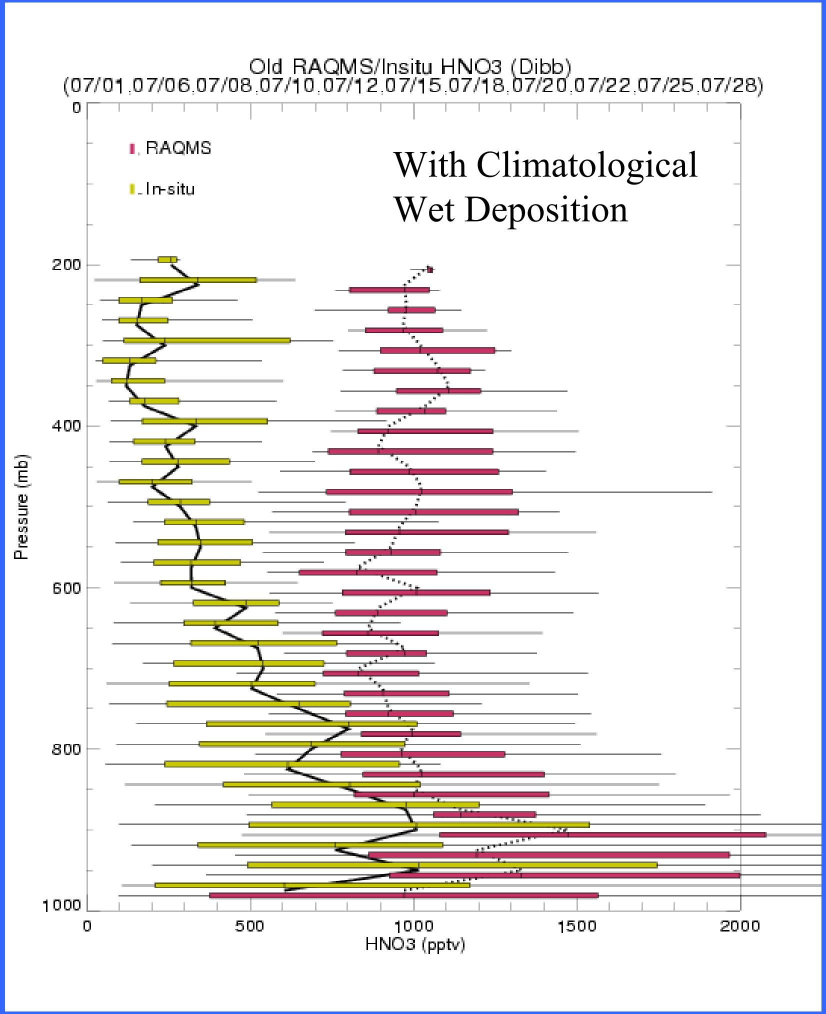
Chemical constraints from 6hr RAQMS global analyses [Pierce, 2003]

EPA Speciated PM2.5 Network 07/20/04



Impact of Improved Wet Deposition on RAQMS Upper Air HNO3: INTEX RAQMS/DC8 Insitu HNO3 (J. Dibb, UNH)



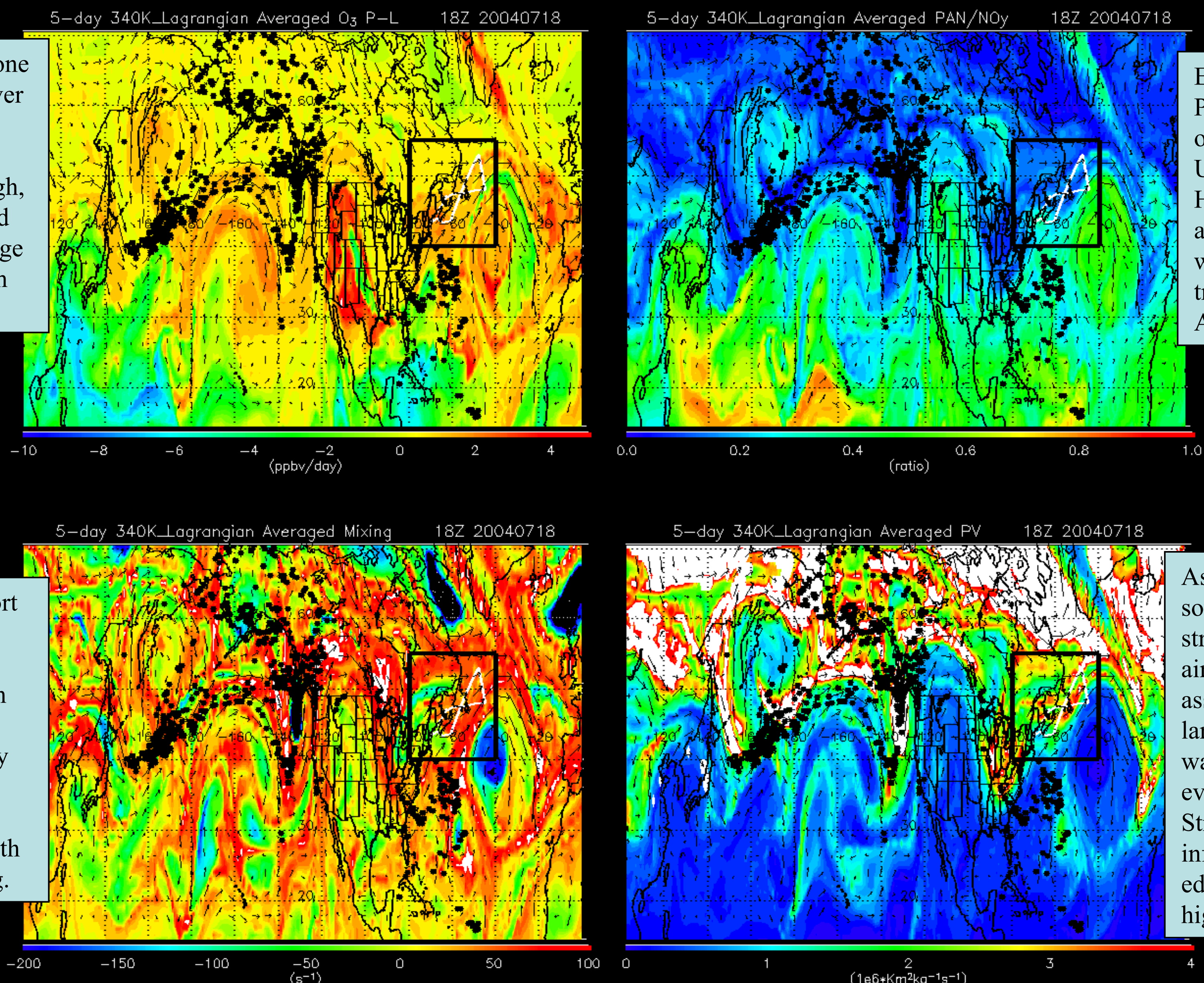


Plan to take advantage of on-line implementation of RAQMS chemistry (instantaneous 3D precipitation, clouds, convective updraft velocities) to improve treatment of wet deposition formulation.

RAQMS$_G$ 340K Lagrangian Analyses 18Z July 18th, 2004

Enhanced ozone production over Western US, spiraling into Bermuda High, and associated with long-range transport from Asia.

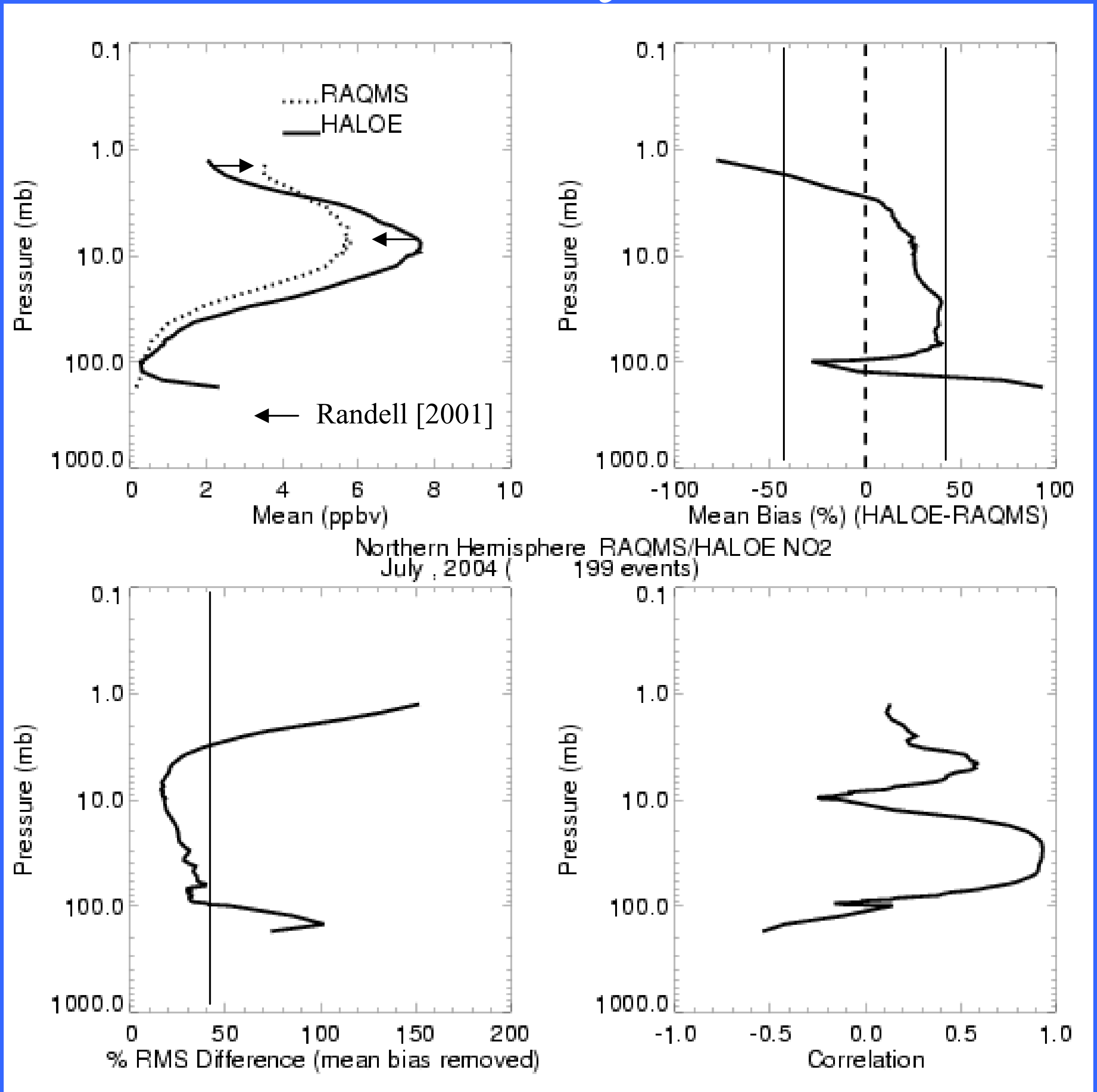
Enhanced PAN/NOy ratios over Western US, Bermuda High, and associated with long-range transport from Asia.



Asian transport within strong mixing zone. Bermuda high shows inner core with very weak mixing (stirring) and outer edge with strong mixing.

Asian transport to south of stratospheric airmass associated with large Rossby wave breaking event. Stratospherically influenced air on edge of Bermuda high.

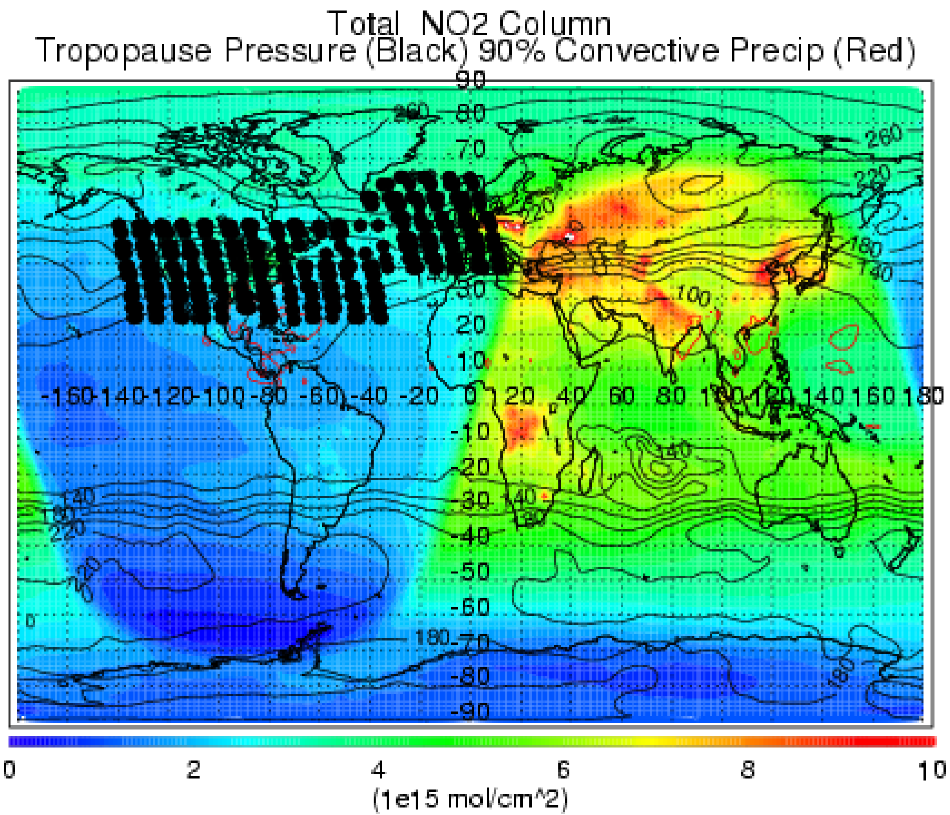
RAQMS vs HALOE NO2 retrieval: July 2004



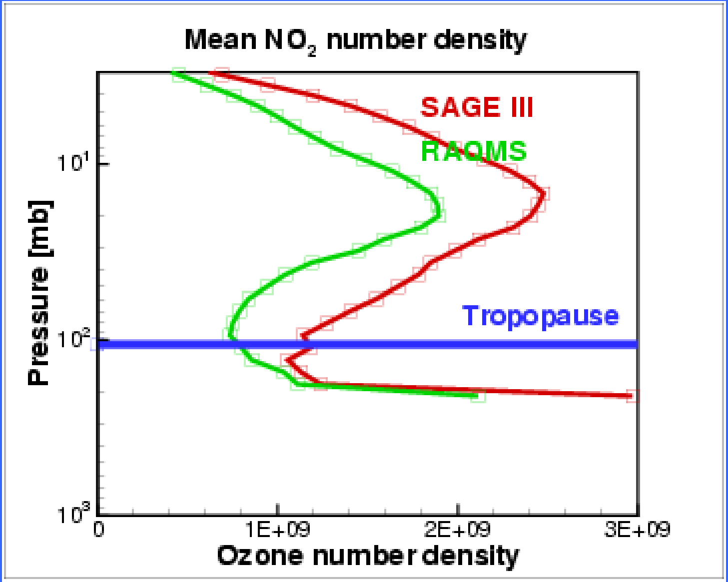
RAQMS NO2 Mean Bias* and RMS Errors < 40% above 100mb.

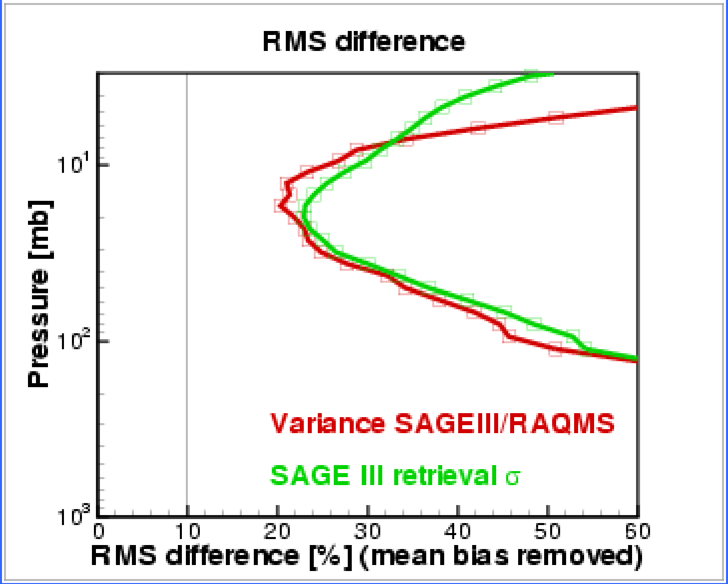
*RAQMS stratospheric NO2 underestimates in middle and lower stratosphere are consistent with systematic NOx/NOy underestimates found during 1997 POLARIS mission [Pierce et al., 1999]

Randell et al. [2001] found HALOE upper stratospheric NO2 low by ~1 ppbv and HALOE middle stratosphierc NO2 high by ~10% relative to POAM.



RAQMS 18Z Total Column NO2 July 2004.





RAQMS vs SAGE III LS NO_2 Mean Bias and RMS Errors are </= Sonde statistics.

RMS Error is consistent with estimated SAGE III LS retrieval uncertainty.